

MERCURY STUDY REPORT TO CONGRESS

VOLUME I:

EXECUTIVE SUMMARY

SAB REVIEW DRAFT

June 1996

**Office of Air Quality Planning and Standards
and
Office of Research and Development**

U.S. Environmental Protection Agency

DEDICATION

The U.S. EPA scientists who authored this report dedicate their efforts to the memory of their colleague, Terry Clark. Terry began his career at the U.S. EPA in 1975, where he became a national, and then an international expert in the atmospheric transport of acid rain and toxic trace gases. Terry designed the initial long-range transport analysis for the Mercury Study. The energy and creativity he brought to his work sustained him even through the final months of his illness when he continued to work daily on this report. His honesty, intelligence and generosity of spirit are greatly missed. Terry Clark died on January 28, 1994.

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LIST OF SYMBOLS, UNITS AND ACRONYMS

| | |
|------------------------------------|--|
| ATSDR | Agency for Toxic Substances and Disease Registry |
| BAF | Bioaccumulation factor |
| bw | Body weight |
| CAA | Clean Air Act as Amended in 1990 |
| CH ₃ Hg | Monomethylmercury |
| (CH ₃) ₂ Hg | Dimethylmercury |
| CSFII | U.S. Department of Agriculture's Continuing Surveys of Individual Food Consumption |
| H ₂ S | Hydrogen sulfide |
| Hg ⁰ | Elemental mercury |
| Hg(II) | Mercuric ion (divalent mercury) |
| HgS | Mercuric sulfide |
| U.S. EPA | U.S. Environmental Protection Agency |
| U.S. FDA | U.S. Food and Drug Administration |
| GACT | Generally available control technology |
| g | Gram |
| HAP | Hazardous Air Pollutant |
| kg | Kilogram (1,000 grams) |
| km | Kilometer (1,000 meters) |
| MACT | Maximum achievable control technology |
| Mg | Megagram (one million grams or one metric ton) |
| ORD | U.S. EPA Office of Research and Development |
| pg | Picogram (10 ⁻¹² gram) |
| ppm | Part per million |
| RfC | Reference concentration |
| RfD | Reference dose |
| µg | Microgram (10 ⁻⁶ gram) |
| WC | Wildlife criterion |
| WHO | World Health Organization |
| WHO/IPCS | World Health Organization's International Programme for Chemical Safety |
| yr | Year |

1. THE MERCURY STUDY REPORT TO CONGRESS

The Clean Air Act Amendments of 1990 (CAA) established section 112(n)(1)(B) which requires the United States Environmental Protection Agency (U.S. EPA) to study the impacts of mercury air pollution. In particular, section 112(n)(B) specifies the following:

The Administrator shall conduct, and transmit to the Congress not later than 4 years after the date of enactment of the Clean Air Act Amendments of 1990, a study of mercury emissions from electric utility steam generating units, municipal waste combustion units, and other sources, including area sources. Such study shall consider the rate and mass of such emissions, the health and environmental effects of such emissions, technologies which are available to control such emissions, and the costs of such technologies.

The U.S. EPA designed the Mercury Study to address many different (but linked) types of information:

- data on type, sources, and trends in emissions;
- evaluation of the atmospheric transport of mercury to locations distant from emission sources;
- assessment of potential impacts of mercury emissions close to the source;
- identification of major pathways of exposure to humans and non-human biota;
- identification of the types of human health consequences of mercury exposure and the amount of exposure likely to result in adverse effects;
- evaluation of mercury exposure consequences for ecosystems and for non-human species;
- identification of populations especially at risk from mercury exposure due to innate sensitivity or high exposure; and
- estimates of control technology efficiencies and costs.

The Report used the above types of information to assess the impact of emissions to air of mercury from a variety of sources. This assessment included judgments as to the potential hazard to humans and wildlife of methylmercury exposure which (as is described in succeeding sections) is largely through the consumption of contaminated fish.

There was no attempt in this Report to do a comparative risk/benefit analysis of fish as an important source of protein and calories in the diet of U.S. populations. Such an analysis would be beyond the scope of the CAA mandate. As emphasized in succeeding sections, the typical U.S. consumer of fish is not in danger of consuming harmful levels of methylmercury and is not being advised to reduce fish consumption.

This Mercury Study Report to Congress fulfills the mandate of section 112(n)(1)(B). The report is in seven volumes:

- Volume I: Executive Summary
- Volume II: An Inventory of Anthropogenic Mercury Emissions in the United States

- Volume III: An Assessment of Exposure from Anthropogenic Mercury Emissions in the United States
- Volume IV: Health Effects of Mercury and Mercury Compounds
- Volume V: An Ecological Assessment of Anthropogenic Mercury Emissions in the United States
- Volume VI: Characterization of Human Health and Wildlife Risks from Anthropogenic Mercury Emissions in the United States
- Volume VII: An Evaluation of Mercury Control Technologies and Costs.

The various analyses documented in this Report were designed and conducted in accordance with accepted guidelines and procedures. For example, the human health risk assessment performed for this Report follows published Guidelines for Risk Assessment (including guidelines on *Exposure Assessment*, *Developmental Toxicity*, *Carcinogenicity* and *Germ Cell Mutagenicity*) and uses established methodologies for quantitative assessment of general systemic toxicity (e.g., in the calculation of reference doses (RfDs) and reference concentrations (RfCs)). Moreover, the assessment of ecological effects, presented in Volume V, follows U.S. EPA's *Framework for Ecological Risk Assessment*. Criteria values for protection of piscivorous wildlife were developed using the methodology developed for the *Great Lakes Water Quality Initiative*.

In 1994, the National Research Council of the National Academy of Sciences, in *Science and Judgment in Risk Assessment*, recommended several areas in which U.S. EPA could improve its risk assessment and risk characterization practices. These recommendations are listed below along with a description of how they were implemented in this Report.

- *Provide an understanding of the type and magnitude of an adverse effect that a specific chemical or emission could cause under particular circumstances.* The Report characterizes both the type and magnitude of health and ecological effects associated with airborne emissions of mercury from anthropogenic sources.
- *Validate methods and models.* All models used for the Report were critiqued by scientific experts and model predictions were compared to measured mercury levels using the most appropriate data available.
- *Describe the basis for default options.* All assumptions are described and justified based on available data. Where appropriate, exposure models were modified to improve assumptions and to focus on areas of prediction where use of model assumptions is most justified.
- *Articulate and prioritize data needs.* The Report includes a section on Research Needs in each volume.
- *Distinguish between variability and uncertainty.* The Report provides discussions that attempt to make these distinctions for the risk results.
- *Perform formal uncertainty analyses.* Uncertainty analyses were formally conducted for the dose-response and exposure assessment steps of the study, and were implicit in weight-of-evidence processes used in the hazard identification step of the human health risk assessment and the problem formulation phase of the ecological risk assessment. Uncertainty also was analyzed quantitatively in other components of the study, such as in the calculation of bioaccumulation factors and the RfD for methylmercury.

2. MERCURY IN THE ENVIRONMENT

As a chemical element, mercury cannot be created or destroyed. The same amount has existed on the planet since the earth was formed. Mercury, however, can cycle in the environment as part of both natural and human (anthropogenic) activities. Measured data and modeling results indicate that the amount of mercury mobilized and released into the biosphere has increased since the beginning of the industrial age.

Several types of emission sources contribute to the total atmospheric loading of mercury. Once in the air, mercury can be widely dispersed and transported thousands of miles from likely emission sources. Studies indicate that the residence time of mercury in the atmosphere may be on the order of a year, allowing its distribution over long distances, both regionally and globally, before being deposited to the earth. Even after it deposits, mercury commonly is emitted back to the atmosphere either as a gas or in association with particulates to be re-deposited elsewhere. Mercury undergoes a series of complex chemical and physical transformations as it cycles among the atmosphere, land, and water. Humans, plants and animals are routinely exposed to mercury and accumulate it during this cycle, potentially resulting in a variety of ecological and human health impacts.

Properties and Uses of Mercury

Elemental mercury metal is a heavy, silvery-white liquid at typical ambient temperatures and atmospheric pressures. The vapor pressure of mercury metal is strongly dependent on temperature, and it vaporizes readily under ambient conditions. Most of the mercury encountered in the atmosphere is elemental mercury vapor.

Mercury can exist in three oxidation states: Hg^0 (metallic), Hg_2^{2+} (mercurous) and Hg^{2+} (mercuric). The properties and behavior of mercury depend on the oxidation state. Most of the mercury in water, soil, sediments, or biota (i.e., all environmental media except the atmosphere) is in the form of inorganic mercury salts and organic forms of mercury.

Mercury is widely used because of its diverse properties. In very small quantities, mercury conducts electricity, responds to temperature and pressure changes and forms alloys with almost all other metals. Mercury serves an important role as a process or product ingredient in several industrial sectors.

In the electrical industry, mercury is used in components such as fluorescent lamps, wiring devices and switches (e.g., thermostats) and mercuric oxide batteries. Mercury also is used in navigational devices, instruments that measure temperature and pressure and other related uses. It also is a component of dental amalgams used in repairing dental caries (cavities).

In addition to specific products, mercury is used in numerous industrial processes. The largest quantity of mercury used in manufacturing in the U.S. is the production of chlorine and caustic soda by mercury cell chlor-alkali plants. Other processes include amalgamation, use in nuclear reactors, wood processing (as an anti-fungal agent), use as a solvent for reactive and precious metals, and use as a catalyst. Mercury compounds are also frequently added as a preservative to many pharmaceutical products.

The Role of Atmospheric Releases and Processes

A schematic of the most recent conceptualization of the current global mercury cycle is presented in Figure 2-1. As indicated in this figure, mercury is emitted to the atmosphere by a variety of sources, dispersed and transported in the air, deposited to the earth, and stored in or transferred between the land, water, and air.

Figure 2-1
The Global Mercury Cycle

[Figure 1 from page 2 of the Expert Panel Report]

Source: Adapted from Mason, R.P., Fitzgerald, W.F., and Morel, M.M. 1994. The Biogeochemical Cycling of Elemental Mercury: Anthropogenic Influences. *Geochim. Cosmochim. Acta*, in press.

Mercury deposits on the earth in different ways and at different rates, depending on its physical and chemical form. Mercuric species are subject to much faster atmospheric removal than elemental mercury. Mercuric mercury bound to airborne particles and in a gaseous form is readily scavenged by precipitation and is also dry deposited (that is, deposited in the absence of precipitation). In contrast, elemental mercury vapor has a strong tendency to remain airborne and is not susceptible to any major process resulting in direct deposition to the earth's surface. Although much uncertainty still exists, several studies indicate that the relative contribution of mercury loadings to land and water from atmospheric deposition can be substantial.

Numerous studies of elevated mercury levels in remote locations, where atmospheric transport and deposition appears to be the primary mechanism for contamination, provide further evidence of the importance of the atmospheric pathway.

Fate and Transport of Mercury in the Environment

The movement and distribution of mercury in the environment can be confidently described only in general terms. There has been increasing consensus on many, but not all, of the detailed behaviors of mercury in the environment. The depiction of the mercury cycle in Figure 2-2 illustrates the major transfer and transformation processes expected to occur. These processes include a number of infinite and/or indefinite loops.

Figure 2-2 Cycling of Mercury in Freshwater Lakes

[Figure 2-1 from page 2-3 of the 12/13/94 version of Volume V]

Source: Adapted from Winfrey, M.R. and J.W.M. Rudd. 1990. Review -- Environmental Factors Affecting the Formation of Methylmercury in Low pH Lakes. *Environ. Toxicol. Chem.* 9:853-869.

Mercury cycling and partitioning in the environment are complex phenomena that depend on numerous environmental parameters. The following points generally describe the key factors that affect the fate and transport of mercury in the environment.

- The form of mercury in air affects both the rate and mechanism by which it deposits to earth.
- Wet deposition apparently is the primary mechanism for transporting mercury from the atmosphere to surface waters and land.
- Once in aquatic systems, mercury can exist in dissolved or particulate forms and can undergo a number of chemical transformations (see Figure 2-2).

- Contaminated sediments at the bottom of surface waters can serve as an important mercury reservoir, with sediment-bound mercury recycling back into the aquatic ecosystem for decades or longer.
- Mercury has a long retention time in soils. As a result, mercury that has accumulated in soils may continue to be released to surface waters and other media for long periods of time, possibly hundreds of years.

Potential Exposure Pathways

Plants, animals and humans can be exposed to mercury by direct contact with contaminated environmental media or ingestion of mercury-contaminated water and food.

Generally, mercury accumulates up aquatic food chains so that organisms in higher trophic levels have higher mercury concentrations. An example aquatic food web is shown in Figure 2-3. At the top trophic levels are piscivores, such as humans, bald eagles, cormorants, herring gulls and other fish-eating species. The larger wildlife species (e.g., bald eagle, otter) can prey on fish that occupy high trophic levels, such as trout and salmon, which in turn feed on smaller "forage" fish. Smaller piscivorous wildlife (e.g., kingfishers, ospreys) tend to feed on the smaller forage fish, which in turn feed on zooplankton or benthic invertebrates. Zooplankton feed on phytoplankton and the smaller benthic invertebrates feed on algae and detritus. Thus, mercury is transferred and accumulated through several trophic levels.

Figure 2-3
Example Aquatic Food Web

[Newly drawn figure to be pasted in]

Mercury Methylation and Bioaccumulation

Methylation of mercury is a key step in the entrance of mercury into food chains. The biotransformation of inorganic mercury species to methylated organic species in water bodies can occur in the sediment and the water column. All mercury compounds entering an aquatic ecosystem, however, are not methylated; demethylation reactions as well as volatilization of dimethylmercury decrease the amount of methylmercury available in the aquatic environment. There is a large degree of scientific uncertainty regarding the rate at which these reactions take place. There is general scientific agreement however that there is significant variability between waterbodies concerning the environmental factors that influence the methylation of mercury.

Nearly 100% of the mercury that bioaccumulates in fish tissue is methylated. A relationship exists between the methylmercury content in fish and lake pH, with higher methylmercury content in fish tissue typically found in more acidic lakes. Numerous factors in addition to low pH can influence the bioaccumulation of mercury in aquatic biota. These include the length of the aquatic food chain, temperature and dissolved organic material. Physical and chemical characteristics of a watershed affect the amount of mercury that is translocated from soils to water bodies. Interrelationships between these factors are poorly understood, however, and there is no single factor (including pH) that has been correlated with mercury bioaccumulation in all cases examined.

Mercury accumulates in an organism when the rate of uptake exceeds the rate of elimination. Although all forms of mercury can accumulate to some degree, methylmercury generally accumulates to a greater extent than other forms of mercury. Inorganic mercury can also be absorbed but is generally taken up at a slower rate and with lower efficiency than is methylmercury. Elimination of methylmercury takes place very slowly resulting in tissue half-lives (i.e., the time in which half of the mercury in the tissue is eliminated) ranging from months to years. Elimination of methylmercury from fish is so slow that long-term reductions of mercury concentrations in fish are often due mainly to growth of the fish. In comparison, other mercury compounds are eliminated relatively quickly resulting in reduced levels of accumulation.

Methylmercury production and accumulation in the freshwater ecosystem is an efficient process for accumulating mercury which can then be ingested by piscivores including birds, non-human mammals and people. In addition, methylmercury generally comprises a relatively greater percentage of the total mercury content at higher trophic levels. Accordingly, mercury exposure and accumulation is of particular concern for animals at the highest trophic levels in aquatic food webs and for animals and humans that feed on these organisms.

Human Exposure Pathways and Health Effects

Humans are most likely to be exposed to methylmercury through fish consumption. Exposure may occur through other routes as well (e.g., the ingestion of methylmercury-contaminated drinking water and food sources other than fish, and dermal uptake through soil and water); however, the fish consumption pathway dominates these other pathways for people who eat fish.

There is a great deal of variability among individuals in fish-eating populations with respect to food sources and fish consumption rates. The populations most highly exposed are those located in areas where the concentration of methylmercury in freshwater fish is elevated, in part as the result of anthropogenic releases from industrial and combustion sources. Methylmercury exposure rates among children who consume fish are predicted to be higher than for adults who consume fish because of their lower body weight. Humans could also be exposed to inorganic mercury through inhalation, or

consumption of contaminated water or food. Inhalation exposure to elemental mercury is largely confined to humans whose occupations put them in contact with elemental mercury vapors.

Mercury is a known human toxicant which has been associated with occupational exposure (for example, “Mad Hatters’ Disease”) and with exposure through consumption of contaminated food. Studies in humans and in experimental animals are described in Volume IV of the Mercury Study Report to Congress. Generally, the effect seen at the lowest exposure level for elemental and methylmercury is neurotoxicity. The range of neurotoxic effects can vary from subtle decrements in motor skills and sensory ability to tremors, inability to walk, convulsions and death.

Environmental Impacts

Effects of mercury on fish include death, reduced reproductive success, impaired growth and development and behavioral abnormalities. Exposure to mercury can also cause adverse effects in plants, birds and mammals. Reproductive effects are the primary concern for avian mercury poisoning and can occur at dietary concentrations well below those which cause overt toxicity. Sublethal effects of mercury on birds include liver damage, kidney damage, and neurobehavioral effects. Effects of mercury on plants include death and sublethal effects. Sublethal effects on aquatic plants can include plant senescence, growth inhibition and decreased chlorophyll content. Sublethal effects on terrestrial plants can include decreased growth, leaf injury, root damage, and inhibited root growth and function.

Although clear causal links between mercury contamination and population declines in various wildlife species have not been established, mercury may be a contributing factor to population declines of the endangered Florida panther and the common loon. Some researchers have concluded, however, that mercury levels in most areas are not high enough to adversely affect bird populations.

The National Mercury Problem

Current levels of mercury in freshwater fish in the United States are such that advisories have been issued in 35 states that warn against the consumption of certain amounts and species of fish that are contaminated with mercury. Six states (based on 1994 data) have statewide advisories (i.e., advisories posted on every freshwater body in that state). These advisories are based on the results of sampling surveys that measure mercury levels in representative fish species collected from water bodies. The advisories are intended for people who catch or eat fish from those waterbodies. The States have the discretion of establishing action levels which are different from those of the FDA.

Fish in commerce are under the jurisdiction of the FDA which issues action levels for concentration of mercury in fish and shellfish. The current action level is 1 ppm mercury based on a consideration of health impacts. The typical U.S. consumer of commercial seafood is not in danger of ingesting harmful levels of methylmercury in seafood. The existing FDA management and consumer advice is protective of food in commerce. In some areas, freshwater fish can have mercury levels which exceed the U.S. FDA action limit of 1 ppm. The concentration of methylmercury in commercially important marine species is, on the average, lower than the FDA action level.

3. FINDINGS OF THE MERCURY STUDY REPORT TO CONGRESS

Sources Contributing to Mercury in the Environment

In the CAA, Congress directed U.S. EPA to examine sources of mercury emissions, including electric utility steam generating units, municipal waste combustion units and other sources, including area sources. The U.S. EPA interpreted the phrase "... and other sources..." to mean that a comprehensive examination of mercury sources should be made and to the extent data were available, air emissions should be quantified. Volume II of this report describes in some detail various source categories that emit mercury. In many cases, a particular source category is identified as having the potential to emit mercury, but data are not available to assign a quantitative estimate of emissions. The U.S. EPA's intent was to identify as many sources of mercury emissions to the air as possible and to quantify those emissions where possible.

The mercury emissions data that are available vary considerably in quantity and quality among different source types. Not surprisingly, the best available data are for source categories that U.S. EPA has examined in the past or is currently studying.

Sources of mercury emissions in the United States are ubiquitous. To characterize these emissions, the types are defined in the following way:

- *Natural mercury emissions* -- the mobilization or release of geologically bound mercury by natural processes, with mass transfer of mercury to the atmosphere;
- *Anthropogenic mercury emissions* -- the mobilization or release of geologically bound mercury by human activities, with mass transfer of mercury to the atmosphere; or
- *Re-emitted mercury* -- the mass transfer of mercury to the atmosphere by biologic and geologic processes drawing on a pool of mercury that was deposited to the earth's surface after initial mobilization by either anthropogenic or natural activities.

Contemporary anthropogenic emissions of mercury are only one component of the global mercury cycle. Releases from human activities today are adding to the mercury reservoirs that already exist in land, water, and air, both naturally and as a result of previous human activities. Given the present understanding of the global mercury cycle, the flux of mercury from the atmosphere to land or water at any one location is comprised of contributions from the following:

- The natural global cycle,
- The global cycle perturbed by human activities,
- Regional sources, and
- Local sources.

Local sources could also include direct water discharges in addition to air emissions. Past uses of mercury, such as fungicide application to crops are also a component of the present mercury burden in the environment.

Understanding of the global mercury cycle (shown schematically in Figure 3-1) has improved significantly with continuing study of source emissions, mercury fluxes to the earth's surface, and the

Figure 3-1
Comparison of Current and Pre-Industrial
Mercury Budgets and Fluxes

[Figures shown in Appendix B, page 19 of the Expert Panel Report]

Source: Adapted from Mason, R.P. Fitzgerald, W.F. and Morel, M.M. 1994. The Biogeochemical Cycling of Elemental Mercury: Anthropogenic Influences. *Geochem. Cosmochim. Acta*, in press.

magnitude of mercury reservoirs that have accumulated in soils, watersheds and ocean waters. Although considerable uncertainty still exists, it has become increasingly evident that anthropogenic emissions of mercury to the air rival or exceed natural inputs. Recent estimates place the annual amounts of mercury released into the air by human activities at between 50 and 75 percent of the total yearly input to the atmosphere from all sources. Recycling of mercury at the earth's surface, especially from the oceans, extends the influence and active lifetime of anthropogenic mercury releases.

A better understanding of the relative contribution of mercury from anthropogenic sources is limited by substantial remaining uncertainties regarding the level of natural emissions as well as the amount and original source of mercury that is re-emitted to the atmosphere from existing reservoirs. Recent estimates indicate that of the approximately 200,000 tons of mercury emitted to the atmosphere since 1890, about 95 percent resides in terrestrial soils, about 3 percent in the ocean surface waters, and 2 percent in the atmosphere. More study is needed before it is possible to accurately differentiate natural fluxes from these reservoirs from re-emissions of mercury originally released from anthropogenic sources. For instance, approximately one-third of total current global mercury emissions are thought to cycle from the oceans to the atmosphere and back again to the oceans, but a major fraction of the emissions from oceans consists of recycled anthropogenic mercury. It is believed that much less than 50 percent of the oceanic emission is from mercury originally mobilized by natural sources. Similarly, a potentially large fraction of terrestrial and vegetative emissions consists of recycled mercury from previously deposited anthropogenic and natural emissions.

Comparisons of contemporary (within the last 15-20 years) measurements and historical records indicate that the total global atmospheric mercury burden has increased since the beginning of the industrialized period by a factor of between two and five (see Figure 3-1). For example, analysis of sediments from Swedish lakes shows mercury concentrations in the upper layers that are two to five times higher than those associated with pre-industrialized times. In Minnesota and Wisconsin, an investigation of whole-lake mercury accumulation indicates that the annual deposition of atmospheric mercury has increased by a factor of three to four since pre-industrial times. Similar increases have been noted in other studies of lake and peat cores from this region, and results from remote lakes in southeast Alaska also show an increase, though somewhat lower than found in the upper midwest U.S.

While the overall trend in the global mercury burden since pre-industrial times appears to be increasing, there is some evidence that mercury concentrations in the environment in certain locations have been stable or decreasing over the past few decades. For example, preliminary results for eastern red cedar growing near industrial sources (chlor-alkali, nuclear weapons production) show peak mercury concentrations in wood formed in the 1950s and 1960s, with stable or decreasing concentrations in the past decade. Some results from peat cores and lake sediment cores also suggest that peak mercury deposition occurred prior to 1970. Data collected over 25 years from many locations in the United Kingdom on liver mercury concentrations in two raptor species and a fish-eating grey heron indicate that peak concentrations occurred prior to 1970. The sharp decline in liver mercury concentrations in the early 1970s suggests that local sources, such as agricultural uses of fungicides, may have led to elevated mercury levels two to three decades ago. Similar trends have been noted for mercury levels in eggs of the common loon collected from New York and New Hampshire. This downward trend in mercury concentrations observed in the environment over the last few decades generally tracks with mercury use and consumption patterns over the same timeframe (discussed below).

While selected studies provide some evidence of declining mercury concentrations on a very localized level, there does not appear to be a decrease in the global mercury burden. For example, a

1992 national study by U.S. EPA found mercury residues in fish at 92 percent of more than 314 surface water bodies tested in the U.S.. This study found mercury levels above 1 part per million (ppm), the level used by the Food and Drug Administration as a basis for banning the sale of fish, at 2 percent of the sites surveyed, and above 0.5 ppm, a consumption advisory level used in many states, at 15 percent of the sites surveyed. More recent and complete data are not available to evaluate the current trend in mercury concentrations in fish tissue. As of 1994, however, sixty percent of the almost 1,300 consumption advisories issued in the U.S. due to toxic contamination in fish were for mercury contamination. As of 1994, 35 states had at least one waterbody under mercury advisory, including six states with statewide mercury advisories.

Given the considerable uncertainties regarding the levels of natural and re-emitted mercury emissions, the emissions inventory focused only on the nature and magnitude of mercury emissions from anthropogenic sources. The U.S. EPA recognizes, however, that an assessment of the relative public health and environmental impact that can be attributed to current anthropogenic emissions is greatly complicated by both natural mercury emissions, previous emissions of mercury that have subsequently deposited and other sources such as water discharges and other previous uses (e.g., fungicide application). Further study is needed to determine the importance of natural and re-emitted mercury, and the contribution of water discharges relative to atmospheric deposition.

Inventory Approach and Uncertainties

For most anthropogenic source categories, an emission factor-based approach was used to develop both facility-specific estimates for modeling purposes and nationwide emission estimates. This approach requires an emission factor, which is a ratio of the mass of mercury emitted to a measure of source activity. It also requires an estimate of the annual nationwide source activity level. Examples of measures of source activity include total heat input for fossil fuel combustion and total raw material used or product generated for industrial processes. Emission factors are generated from emission test data, from engineering analyses based on mass balance techniques, or from transfer of information from comparable emission sources. Emission factors reflect the "typical control" achieved by the air pollution control measures applied across the population of sources within a source category.

The emission factor-based approach does not generate exact emission estimates. Uncertainties are introduced in the estimation of emission factors, control efficiencies and the activity level measures. Ideally, emission factors are based on a substantial quantity of data from sources that represent the source category population. For trace pollutants like mercury, however, emission factors are frequently based on limited data that may not have been collected from representative sources. Changes in processes or emission measurement techniques over time may also result in biased emission factors. Emission control estimates are also generally based on limited data; as such, these estimates are imprecise and may be biased. Further uncertainty in the emission estimates is added by the sources of information used on source activity levels, which vary in reliability.

Once emitted to the environment, the fate and transport of mercury is greatly influenced by the chemical form of mercury. The data collected for the emissions inventory was essentially all reported as total mercury with the exception of utility boilers for which there were limited speciated samples. In the exposure analysis described below, estimates were made of speciation profiles for modeling purposes.

To improve the emissions estimates, a variety of research activities are needed. These are listed in Chapter 5 of this volume.

Anthropogenic Emissions Summary

Table 3-1 summarizes the estimated national mercury emission rates by source category. While these emission estimates for anthropogenic sources have limitations, they do provide insight into the relative magnitude of emissions from different groups of sources. All of these emissions estimates should be regarded as best point estimates given available data.

Of the estimated 220 Megagrams (Mg) (243 tons) of mercury emitted annually into the atmosphere by anthropogenic sources in the United States, approximately 85 percent is from combustion point sources, 13 percent is from manufacturing point sources, 1 percent is from miscellaneous sources and 1 percent is from area sources. Four specific source categories account for approximately 83 percent of the total anthropogenic emissions -- medical waste incineration (27 percent), municipal waste combustion (23 percent), utility boilers (21 percent), and commercial/ industrial boilers (12 percent). It should be noted that the U.S. EPA has finalized mercury emission limits for municipal waste combustors, and has proposed mercury emission limits for medical waste incinerators. These emission limits will reduce mercury emissions from these sources by 90 percent.

All four of the most significant sources represent high temperature waste combustion or fossil fuel processes. For each of these operations, the mercury is present as a trace contaminant in the fuel or feedstock. Because of its relatively low boiling point, mercury is volatilized during high temperature operations and discharged to the atmosphere with the exhaust gas.

Trends in Mercury Emissions

It is difficult to predict with certainty the temporal trends in mercury emissions for the U.S., although there appears to be a trend toward decreasing total mercury emissions from 1990 to 1995. This is particularly true for the combustion sources wherein mercury is a trace contaminant of the fuel. Also, as previously noted, there are a number of source categories where there is insufficient data to estimate current emissions let alone potential future emissions. Based on available information, however, a number of observations can be made regarding mercury emission trends from source categories where some information is available about past activities and projected future activities.

There has been a real success in the U.S. in the dramatic drop in mercury emissions from manufacturing over the past decade. Current emissions of mercury from manufacturing sources are generally low (with the exception of chlor-alkali plants using the mercury cell process). The emissions of mercury are more likely to occur when the product is broken or discarded. Therefore, in terms of emission trends, one would expect that if the future consumption of mercury remains consistent with the 1993 consumption rate, emissions from most manufacturing sources would remain about the same.

For industrial or manufacturing sources that use mercury in products or processes, the overall consumption of mercury is generally declining. Industrial consumption of mercury has declined by about two thirds between 1988 (1508 Mg) and 1993 (558 Mg). Much of this decline can be attributed to the elimination of mercury as a paint additive (20 percent) and the reduction of mercury in batteries (36 percent). Use of mercury by other source categories remained about the same between 1988 and 1993.

Secondary production of mercury (i.e., recovering mercury from waste products) has increased significantly over the past few years. Of the 558 Mg of mercury used in industrial processes in 1993, 63 percent was provided by secondary mercury producers. This is a two-fold increase since 1991. The number of secondary mercury producers is expected to increase as more facilities open to recover

Table ES-3
Best Point Estimates of National Mercury Emission Rates by Category

| Source of mercury ^a | 1990-1993 Mg/yr ^{b,c} | 1990-1993 tons/yr ^{b,c} | % of Total Inventory |
|---|-----------------------------------|-------------------------------------|-------------------------|
| Area sources | 2.8 | 3.1 | 1.3 |
| Lamp breakage | 1.4 | 1.5 | 0.6 |
| General lab use | 0.7 | 0.8 | 0.3 |
| Dental prep and use | 0.7 | 0.8 | 0.3 |
| Mobile sources | d | d | d |
| Paint use | e | e | e |
| Agricultural burning | d | d | d |
| Landfills | d | d | d |
| Point sources | 217.3 | 239.4 | 98.7 |
| Combustion sources | 186.9 | 205.9 | 84.9 |
| MWIs ^f | 58.8 | 64.7 | 26.7 |
| MWCs | 50 | 55 | 22.7 |
| Utility boilers | 46.5 | 51.3 | 21.2 |
| Coal | (46.3) ^g | (51) | (21.0) |
| Oil | (0.23) | (0.25) | (0.1) |
| Natural gas | (0.002) | (0.002) | (0.0) |
| Commercial/industrial boilers | 26.3 | 29 | 12.0 |
| Coal | (20.7) | (22.8) | (9.4) |
| Oil | (5.5) | (6.0) | (2.5) |
| Residential boilers | 3.2 | 3.5 | 1.4 |
| Coal | (0.5) | (0.6) | 0.2 |
| Oil | (2.7) | (3.0) | (1.2) |
| SSIs | 1.7 | 1.8 | 0.7 |
| Crematories | 0.4 | 0.4 | 0.2 |
| Wood-fired boilers ^h | 0.3 | 0.3 | 0.1 |
| Hazardous waste combustors ⁱ | d | d | d |
| Manufacturing sources | 29.1 | 32 | 13.2 |
| Primary lead | 8.2 | 9.0 | 3.7 |
| Secondary Hg production | 6.7 | 7.4 | 3.1 |
| Chlor-alkali | 5.9 | 6.5 | 2.7 |
| Portland cement | 5.9 | 6.5 | 2.7 |
| Primary copper ^j | 0.6 | 0.7 | 0.3 |
| Lime manufacturing | 0.6 | 0.7 | 0.3 |
| Electrical apparatus | 0.42 | 0.46 | 0.2 |
| Instruments | 0.5 | 0.5 | 0.2 |
| Carbon black | 0.23 | 0.25 | 0.1 |
| Fluorescent lamp recycling | 0.005 | 0.006 | 0.002 |
| Batteries | 0.02 | 0.02 | 0.0 |
| Primary Hg production | d | d | d |
| Mercury compounds | d | d | d |
| Byproduct coke | d | d | d |
| Refineries | d | d | d |
| Miscellaneous sources | 1.3 | 1.4 | 0.6 |
| Geothermal power | 1.3 | 1.4 | 0.6 |
| Turf products | e | e | e |
| Pigments, oil, etc. | e | e | e |
| TOTAL | 220.1 | 242.5 | 100.0 |

^a MWC = Municipal waste combustor; MWI = medical waste incinerator; SSI = sewage sludge incinerator.

^b Numbers do not add exactly because of rounding.

^c Where available, emissions estimates for 1995 are discussed in the text. However, these 1995 estimates were not used in any of the modeling analyses.

^d Insufficient information to estimate 1990 emissions.

^e Mercury has been phased out of use.

^f In the course of an MWI rulemaking, with the receipt of new data, U.S. EPA expects to revise the mercury emission estimate for MWIs downward.

^g Parentheses denote subtotal within a larger point source category.

^h Includes boilers only; does not include residential wood combustion (wood stoves).

ⁱ In 1995 incinerators and lightweight aggregate kilns (not cement kilns) were estimated to emit 5.0 tons of mercury.

^j 1990 emissions are estimated for only one source, which ceased operations in February 1995. The nationwide estimate for 1995 is 0.08 tons.

mercury from fluorescent lamps and other mercury-containing products (e.g., thermostats). As a result there is potential for mercury emissions from this source category to increase.

The largest identifiable sources of mercury emissions currently are municipal waste combustors and medical waste incinerators. Emissions from these source categories are expected to decline significantly by the year 2000 due to regulatory action the U.S. EPA is taking under the statutory authority of section 129 of the CAA. The U.S. EPA has finalized rules for municipal waste combustors and proposed rules for medical waste incinerators that will reduce mercury emissions from both of these source categories by about 90 percent. In addition to this federal action, a number of states (including Minnesota, Florida and New Jersey) have implemented mandatory recycling programs to reduce mercury-containing waste, and some states have regulations that impose emission limits that are lower than the federal regulation. These factors will reduce national mercury emissions from these source categories even further.

After municipal solid waste and medical waste incinerators have been controlled, the largest remaining identified source of mercury emissions will be fossil fuel combustion by utility boilers, particularly coal combustion. Future trends in mercury emissions from this source category are largely dependent on both the nation's future energy needs and the fuel chosen to meet those needs. Another factor is the nature of actions the utility industry may take in the future to meet air quality requirements under the Clean Air Act.

Trends in Mercury Consumption

Data on industrial demand for mercury show a general decline in domestic mercury use since demand peaked in 1964. Domestic demand fell by 74 percent between 1980 and 1993, and by more than 50 percent since 1988. The rate of decline, however, has slowed since 1990. Further evidence of the declining need for mercury in the U.S. is provided by the general decline in imports since 1988 and the fact that exports have exceeded imports since at least 1989. Approximately 78 percent of the net U.S. exports of mercury during the last five years has come from federal sales, with a steadily increasing portion of the federal sales coming from the National Defense Stockpile managed by DLA. Federal sales accounted for 97 percent of the U.S. demand in 1993.

Most recently, there has been a sharp drop in Federal sales. In July 1994, DLA suspended future sales of mercury from the Department of Defense stockpile until the environmental implications of these sales are addressed. In addition, in past years, DLA sold mercury accumulated and held by the Department of Energy, which is also considered excess to government needs. DLA suspended these mercury sales in July 1993 for an indefinite period in order to concentrate on selling material from its own mercury stockpile.

In general, these data suggest that industrial manufacturers that use mercury are shifting away from mercury except for uses for which mercury is considered essential. This shift is believed to be largely the result of Federal bans on mercury additives in paint and pesticides; industry efforts to reduce mercury in batteries; increasing state regulation of mercury emissions sources and mercury in products; and state-mandated recycling programs. A number of Federal activities are also underway to investigate pollution prevention measures and control techniques for a number of sources categories (see Volume VII of this Report to Congress).

Assessment of Exposure

Exposure Assessment Approach

The exposure assessment draws upon the available scientific information and presents quantitative modeling analyses which examine the following: (1) the long range transport of mercury from emission sources through the atmosphere; (2) the transport of mercury from emission sources through the local atmosphere; (3) the aquatic and terrestrial fate and transport of mercury at hypothetical sites; and (4) finally, the resulting exposures to hypothetical humans and animals that inhabit these sites. Exposure to mercury from seafood was estimated using a cross sectional survey with a three day sampling period and central tendency estimates of mercury concentrations in the tissues of seafood.

There are no data that conclusively demonstrate a relationship between anthropogenic sources and increased mercury concentrations in environmental media or biota. Available mercury monitoring data around sources are extremely limited and no comprehensive database describing environmental concentrations has been developed. To determine if there is a connection between anthropogenic emission sources and increased environmental levels, the exposure assessment in this Report utilized exposure modeling techniques.

Figure 3-2 illustrates the how the various exposure models were integrated to estimate both human and wildlife exposure.

Long-Range Transport Analysis

The long range transport modeling predicts the regional and national deposition of mercury across the continental U.S. Details of several studies which demonstrate the long range transport of mercury are presented in Volume III. In this Report, the long range transport of mercury was modeled using site-specific, anthropogenic emission source data (presented in Volume II of this Report) to generate mean, annual atmospheric mercury concentrations and deposition values across the continental U.S. The Regional Lagrangian Model of Air Pollution (RELMAP) was utilized to model annual mercury emissions from multiple mercury emission sources.

The chemical form of emitted mercury is a critical factor in its fate, transport and toxicity in the environment. With the exception of utility boilers, for which there are limited speciated data, mercury emissions are reported as total measured mercury. The form distributions, or speciation factors, define the estimated fraction of mercury emitted as elemental mercury (Hg^0), divalent mercury (Hg^{2+}), or mercury associated with particulates (Hg_p) were adopted from Peterson et al. (1995). Since there is considerable uncertainty about the speciation profiles, an alternate speciation scenario was also modeled to measure the sensitivity of the RELMAP results to this uncertainty. The speciation factors for the base case and alternate scenario are discussed in Volume III of this Report. The results of the modeling using the base case speciation scenario are described below.

From the RELMAP analysis and a review of field measurement studies, it is concluded that mercury deposition appears to be ubiquitous across the continental U.S., and at, or above, detection limits when measured with current analytic methods. The southern Great Lakes and Ohio River Valley, the Northeast, and scattered areas in the South (particularly in the Miami and Tampa areas) are predicted to have the highest annual rate of deposition of total mercury (above the levels predicted at the 90th percentile). Figure 3-3 illustrates the pattern of mercury deposition across the U.S. This figure also illustrates the boundaries of the RELMAP modeling domain. Measured deposition

Figure 3-2
Fate, Transport and Exposure Modeling Conducted in the Combined COMPDEP and RELMAP Local Impact Analysis

[Figure 1-4 from Vol. III]

Figure 3-3
Total Mercury Wet + Dry Deposition (Base Case)
Units: g/m²

[Figure 5-18 from Vol. III]

estimates are limited, but are available for certain geographic regions. The data that are available corroborate the RELMAP modeling predictions for specific areas. These comparisons are discussed in detail in Volume III.

A wide range of mercury deposition rates is predicted across the continental U.S. The highest predicted rates (i.e., above 90th percentile) are more than 50 times higher than the lowest predicted rates (i.e., below the 10th percentile). The three principal factors that contribute to these modeled and observed deposition patterns are the emission source locations, amount of divalent and particulate mercury emitted or formed in the atmosphere; and climate and meteorology. A facility located in a humid climate is predicted to have a higher annual rate of mercury deposition than a facility located in an arid climate. The critical variables are the estimated washout ratios of elemental and divalent mercury, as well as the annual amount of precipitation. Precipitation is important because it removes various forms of mercury from the atmosphere and deposits them to the surface of the earth.

Mass Balances of Mercury within the Long-range Model Domain

The general mass balance of elemental mercury gas, divalent mercury gas, and particle-bound mercury from the RELMAP simulation results using specified speciation profiles are shown in Table 3-2. Using the meteorologic data from the year 1989, the mass balance shows a total of 223.8 metric tons of mercury emitted to the atmosphere from anthropogenic sources. (This simulated emission total differs from the national totals indicated in Volume II since the states of Alaska and Hawaii are not within the model domain and latex paint emissions are not considered.) The simulation indicates that 77.9 metric tons of anthropogenic mercury emissions are deposited within the model domain and 0.6 metric tones remain in the air within the model domain at the end of the simulation. The remainder, about 145.3 metric tons, is transported outside the model domain and probably diffuses into the global atmospheric reservoir.

Table 3-2
Modeled Mercury Mass Budget in Metric Tons for 1989
Using the Specified Speciation Profiles

| Source/Fate | Hg ^{0a} | Hg ^{2+b} | Hg _p ^c | Total Mercury |
|--|------------------|-------------------|------------------------------|---------------|
| Total U.S. anthropogenic emissions | 92.0 | 92.6 | 39.1 | 223.8 |
| Mass advected from model domain | 90.4 | 29.9 | 25.0 | 145.3 |
| Dry deposited anthropogenic emissions | 0.0 | 39.0 | 0.6 | 39.6 |
| Wet deposited anthropogenic emissions | 1.2 | 23.6 | 13.4 | 38.3 |
| Remaining in air at end of simulation | 0.4 | 0.1 | 0.1 | 0.6 |
| Total deposited anthropogenic emissions | 1.2 | 62.6 | 14.1 | 77.9 |
| Deposition from background Hg ⁰ | 33.0 | 0.0 | 0.0 | 33.0 |
| Mercury deposited from all sources | 34.2 | 62.6 | 14.1 | 111.0 |
| (All figures rounded to the nearest tenth of a metric ton) | | | | |

^a Hg⁰ = Elemental Mercury

^b Hg²⁺ = Divalent Vapor-phase Mercury

^c Hg_p = Particle-Bound/Mercury

The simulation also indicates that 33.0 metric tons of mercury are deposited within the model domain from this global atmospheric reservoir, suggesting that about four times as much mercury is being added to the global reservoir from U.S. emissions as is being deposited from it. The total amount of mercury deposited in the model domain annually from U.S. anthropogenic emissions and from the global background concentration is estimated to be 111.0 metric tons, or about one-half of the total atmospheric emissions from anthropogenic sources in the lower 48 United States.

Of the total anthropogenic mercury mass deposited to the surface in the model domain, 80% is estimated by the RELMAP to come from Hg^{2+} emissions, 18% from Hg_p emissions and 2% from Hg^0 emissions when the base-case emission speciation profiles are used. When the deposition of Hg^0 from the global background is considered in addition to anthropogenic sources in the lower 48 states, the species fractions become 56% Hg^{2+} , 31% Hg^0 and 13% Hg_p . The vast majority of mercury already in the global atmosphere is in the form of Hg^0 and, in general, the anthropogenic Hg^0 emissions do not greatly increase the existing Hg^0 concentration. Although Hg^0 is removed from the atmosphere very slowly, the global background reservoir is large and extraction of mercury from it is significant in terms of the total deposition. It should be noted that dry deposition of Hg^0 is significant only at very high concentrations and has not been included in the RELMAP simulations. Wet deposition is the only major pathway for removal of Hg^0 from the atmosphere. This removal pathway simulated by the RELMAP involves oxidation of mercury by ozone in an aqueous solution; thus, the Hg^0 that is extracted from the atmosphere by the modeled precipitation process would actually be deposited primarily in the form of Hg^{2+} .

Of the 92.0 metric tons of anthropogenic Hg^0 emitted in the lower 48 states, only 1.2 tons (1.3%) is deposited within the model domain, while of the 92.6 metric tons of Hg^{2+} emitted, about 62.6 tons (67.6%) is deposited. Ninety-eight percent of the deposited anthropogenic mercury was emitted in the form of Hg^{2+} or Hg_p . Thus, a strong argument can be made that the combined Hg^{2+} and Hg_p component of anthropogenic mercury emissions can be used as an indicator of eventual deposition of those emissions to the lower 48 states and surrounding areas. The emission inventory and chemical/physical speciations profiles indicate that of all combined Hg^{2+} and Hg_p emissions, about 36% is from medical waste incineration, 30% is from municipal waste combustion, 18% is from electric utility boilers, 11% is from combustion of fossil fuel other than by electric utilities, 1% is from chlor-alkali plants, 1% is from non-ferrous metal smelting, and 2% is from all other sources.

Limitations of the Long-Range Transport (RELMAP) Analysis

There are a number of uncertainties with the RELMAP analysis. These have to do to a large degree with the current state-of-the-science concerning atmospheric chemistry and speciation profiles of mercury emissions. Some of the most important limitations are listed below.

- Comprehensive emissions data for a number anthropogenic and natural sources are not available. This reflects the current developmental nature of emission speciation methods, resulting in few data on the various species of mercury and proportions of vapor and solid forms emitted. Both elemental and divalent mercury species as well as gaseous and particulate forms are known to be emitted from point and area sources.
- Atmospheric chemistry data are incomplete. Some atmospheric reactions of mercury, such as the oxidation of elemental mercury to divalent mercury in cloud water droplets have been reported. Other chemical reactions in the atmosphere that may reduce divalent species to elemental mercury have not been reported.
- There is inadequate information on the atmospheric processes that affect wet and dry deposition of mercury. Atmospheric particulate forms and divalent species of mercury are thought to wet and

dry deposit more rapidly than elemental mercury; however, the relative rates of deposition are uncertain. There is no validated air pollution model that estimates wet and dry deposition of vapor-phase compounds close to the emission source. In addition, there is uncertainty regarding the revolatilization of deposited mercury.

Exposure Assessment of Local Deposition of Mercury

An analysis of the local atmospheric transport of mercury released from anthropogenic emission sources was undertaken to estimate the impacts of mercury from selected, individual sources. Model plants were developed; these are defined as hypothetical facilities that represent actual emissions from existing industrial processes and combustion sources. The model plants were situated in hypothetical locations intended to simulate a site in either the Western or Eastern U.S. This approach was selected because environmental monitoring studies indicate that measured mercury levels in environmental media and biota may be elevated in areas around stationary industrial and combustion sources known to emit mercury. These measured data are detailed in Chapter 2 of Volume III of this Report.

The exposure assessment addressed atmospheric mercury emissions from six combustion and manufacturing source categories: municipal waste combustors (MWCs), medical waste incinerators (MWIs), utility boilers, chlor-alkali plants, primary lead smelters and primary copper smelters. It did not address all anthropogenic emission sources nor did it address emissions from natural sources. In addition, anthropogenic discharges of mercury to waterbodies were not addressed.

The following human exposure routes were included: inhalation, consumption of water, consumption of fish, beef, beef liver, cow's milk, poultry, chicken eggs, pork, lamb, green plants (e.g., leafy vegetables, potatoes, fruits, grains and cereals) and ingestion of soil. Dermal exposures that resulted from contact with soil and water, as well as exposure through inhalation of resuspended dust particles and exposure through the consumption of human breast milk were not evaluated. The only exposure route considered for wildlife was the consumption of freshwater fish.

Atmospheric concentrations and deposition rates were used as inputs to a series of terrestrial and aquatic models described in U.S. EPA's (1990) Methodology for Assessing Health Risks to Indirect Exposure from Combustor Emissions and a 1994 Addendum. The results of these terrestrial and aquatic models were used to predict mercury exposure to hypothetical humans through inhalation, consumption of drinking water and ingestion of soil, farm products (e.g., beef product and vegetables) and fish. These models were also used to predict mercury exposure in hypothetical piscivorous (i.e., fish-eating) birds and mammals through their consumption of fish.

Results of the Exposure Analysis

This exposure analysis, in conjunction with available scientific knowledge, supports a plausible link between mercury emissions from anthropogenic combustion and industrial sources and mercury concentrations in air, soil, water and sediments. The critical variables contributing to this linkage are these:

- the species of mercury that are emitted from the sources, with elemental mercury (Hg^0) mostly contributing to concentrations in ambient air and divalent mercury (Hg^{2+}) mostly contributing to concentrations in soil, water and sediments;
- the overall amount of mercury emitted from a combustion source; and
- the climate conditions.

In addition, this study also supports a plausible link between mercury emissions from anthropogenic combustion and industrial sources and methylmercury concentrations in freshwater fish. The critical variables contributing to this linkage are the following:

- the species of mercury that are emitted, with emitted divalent mercury mostly depositing into local watershed areas and, to a lesser extent the atmospheric conversion of elemental mercury to divalent species which are deposited over greater distances;
- the overall amount of mercury emitted from a source;
- the extent of mercury methylation in the water body; and
- the climate conditions.

From the analysis of deposition and on a comparative basis, the deposition of divalent mercury (Hg^{2+}) close to an emission source is greater for receptors in elevated terrain (i.e., terrain above the elevation of the stack base) than from receptors located in flat terrain (i.e., terrain below the elevation of the stack base). The critical variables are parameters that influence the plume height, primarily the stack height and stack exit gas velocity.

Based on the local scale atmospheric modeling results in flat terrain, at least 75% of the emitted mercury from each facility is predicted to be transported more than 50 km from the facility. The models used in the exposure analysis indicate that, except for utility boilers and intermittent medical waste incinerators, deposition within 10 Km of a facility is generally dominated by emissions from the local source rather than from emissions transported from regional mercury emissions sources.

Consumption of fish is the dominant pathway of exposure to methylmercury for fish-consuming humans and wildlife. There is a great deal of variability among individuals in these populations with respect to food sources and fish consumption rates. As a result, there is a great deal of variability in exposure to methylmercury in these populations. The anthropogenic contribution to the total amount of methylmercury in fish is, in part, the result of anthropogenic mercury releases from industrial and combustion sources increasing mercury body burdens in fish. As a consequence of human and wildlife consumption of the affected fish, there is an incremental increase in exposure to methylmercury. Due to differences in fish consumption rates per body weight and differences in body weights among species, it is likely that piscivorous birds and mammals have much higher environmental exposures to methylmercury than humans through the consumption of contaminated fish. This is true even in the case of fish consumption by humans who consume above average amounts of fish. The critical variables contributing to these outcomes are these:

- the fish consumption rate;
- the body weight of the individual in relation to the fish consumption rate; and
- the rate of biomagnification between trophic levels within the aquatic food-chain.

In terms of methylmercury intake on a per body weight basis, the five wildlife species considered in this analysis can be ranked from high to low as follows:

- Kingfisher
- River Otter
- Mink, Osprey
- Bald eagle

Methylmercury exposures for the most exposed wildlife species (the kingfisher) may be up to two orders of magnitude higher than human exposures from contaminated freshwater fish (on a kilogram fish consumed per body weight basis). This assumes that the fish within different tropic levels of a given lake are contaminated with the same concentrations of methylmercury.

Some Limitations of the Local Exposure Assessment

Limitations of the local exposure assessment include a lack of information concerning the movement of mercury from watershed soils to water bodies. There are not conclusive data on the amount of and rates of mercury methylation in different types of water bodies. In addition, there is a lack of data on the transfer of mercury between environmental compartments and biologic compartments; for example, the link between the amount of mercury in the water body and the levels in fish appears to vary from water body to water body.

On a national scale, an apportionment between sources of mercury and mercury in environmental media and biota cannot be described in quantitative terms with the current scientific understanding of the environmental fate and transport of this pollutant. There is a lack of adequate mercury measurement data near the anthropogenic atmospheric mercury sources considered in this report. To assess how well the modeled data predict actual mercury concentrations in different environmental media at a variety of geographic locations requires a data base against which to make these comparisons. The lack of such measured data preclude a comparison of the modeling results with measured data around these sources. These data include measured mercury deposition rates as well as measured concentrations in the atmosphere, soils, water bodies and biota. Substantial additional monitoring data would facilitate such comparison.

Assessment of Fish Consumption

A current assessment of U.S. general population methylmercury exposure through the consumption of fish is provided in Chapter 3 and in Appendix H of Volume III. This assessment was conducted to provide an estimate of mercury exposure through the consumption of fish to the general U.S. population. It is not a site-specific assessment but rather a national assessment. This assessment utilizes data from the 1989 - 1991 Continuing Surveys of Food Intake by Individuals (CSFII 89-91) to estimate a range of fish consumption rates among U.S. fish eaters. The CSFII is a cross sectional survey with a three day sampling period. The survey was conducted over a period of three years; it included all seasons and both weekend and weekday sampling. Only individuals who reported fish consumption were considered in the analyses in this Report to Congress. For each fish-eater, CSFII 89-91 study identified the number of fish meals, the quantities and species of fish consumed and the self-reported body weights of the consumers. The constitution of the survey population was weighted to reflect the actual U.S. population.

These estimates of fish consumption rates were combined with species-specific mean values for measured methylmercury concentrations. The marine fish methylmercury concentration data were obtained from the National Marine Fisheries Service Database. The freshwater fish methylmercury concentration data were obtained from Bahnick et al., (1994) and Lowe et al., (1985). Through the application of specific fish preparation factors (USDA, 1995), estimates of the range of methylmercury exposure from the consumption of fresh water fish were prepared for the fish-consuming segment of the U.S. population. Per body weight estimates of methylmercury exposure were determined by dividing the total daily methylmercury exposure from this pathway by the self-reported body weights. The results of his analysis show that children on a per kilogram body weight basis have higher average exposure rates to methylmercury through the consumption of fish than adults.

Human Health Effects of Methylmercury

Data in both humans and experimental animals show that all three forms of mercury evaluated in this Report (elemental, inorganic and methylmercury) can produce adverse health effects. Human exposure to elemental mercury occurs in some occupations, and exposure to inorganic mercury can arise from mercury amalgams used in dental restorative materials (U.S. PHS, Environmental Health Policy Committee, 1995). People, however, are primarily exposed to methylmercury in fish. The focus of this assessment, therefore, is on methylmercury, which can produce a variety of adverse effects, depending on the dose and time of exposure.

Individual risk assessors and specific organizations may choose different risk assessment methodologies. Part of these differences occur when identifying populations or subpopulations of concern. More than one approach to selection of the population at risk of adverse effect is feasible. For example, if children are judged to be the subpopulation of greatest concern, specific age-groups within this subpopulation may be judged to be of greater interest; e.g., birth through 4 years of age. Alternatively other risk assessors may prefer to consider all children (e.g., birth through 14 years of age) as a group when evaluating risk to children.

Neurotoxicity is the effect of greatest concern when exposure occurs to the developing embryo/fetus during pregnancy as well as when adults and children are exposed to methylmercury. Two major epidemics of methylmercury poisoning through fish consumption have occurred. The best known of these two epidemics occurred among people and wildlife living near Minamata City on the shores of Minamata Bay, Kyushu, Japan. The source of methylmercury was a chemical factory that used mercury as a catalyst. A series of chemical analyses identified methylmercury in the factory waste sludge, which was drained into Minamata Bay. Once present in Minamata Bay, the methylmercury accumulated in the tissue of shellfish and fish that were subsequently consumed by wildlife and humans. Fish was a routine part of the diet in these populations. An average fish consumption was reported to be in excess of 300 g/day (reviewed by Harada et al., 1995); this is a greater level of fish consumption than is typical for the general U.S. population.

The first poisoning case occurred in 1956 in a 6-year-old girl who came to a hospital complaining of symptoms characteristic of nervous system damage. Symptoms of Minamata disease in children and adults included the following:

- Impairment of the peripheral vision;
- disturbances in sensations ("pins and needles" feelings, numbness) usually in the hands and feet and sometimes around the mouth;
- incoordination of movements as in writing;
- impairment of speech;
- impairment of hearing;
- impairment of walking; and
- mental disturbances.

It frequently took several years before people were aware that they were developing the signs and symptoms of methylmercury poisoning.

Over the next 20 years the number of people known to be affected with what became known as Minamata disease increased to thousands. In time the disease was recognized to result from methylmercury poisoning, and fish were subsequently identified as the source of methylmercury. As is often the situation with epidemics, the first cases noted were severe. Deaths occurred among both adults and children. It also was recognized that the nervous system damage could occur to the fetus if the mother ate fish contaminated with high concentrations of methylmercury during pregnancy. The nervous system damage of severe methylmercury poisoning among infants was very similar to congenital cerebral palsy.

In the fishing villages of this region the occurrence of congenital cerebral palsy due to methylmercury was very high compared to the incidence for Japan in general. After the source of mercury contamination was identified, efforts were made to reduce the release of mercury into the bay. After 1969, average mercury concentrations in fish had fallen below 0.5 ppm.

In 1965, an additional methylmercury poisoning outbreak occurred in the area of Niigata, Japan. As in Minamata, multiple chemical plant sources of the chemical were considered. Scientific detective work identified the source again to be a chemical factory releasing methylmercury into the Agano River. The signs and symptoms of disease in Niigata were those of methylmercury poisoning and strongly similar to the disease in Minamata.

The abnormalities (or pathology) in the human brain that result from methylmercury poisoning are well described. There is an extremely high level of scientific certainty that methylmercury causes these changes. Similar pathology has been identified in other countries where methylmercury poisonings have occurred. Methylmercury contamination of other food products (including grains and pork products) has resulted in severe methylmercury poisoning with pathological changes in the nervous system and clinical disease virtually identical to Minamata disease.

Methylmercury poisoning occurred in Iraq following consumption of seed grain that had been treated with a fungicide containing methylmercury. The first outbreak occurred prior to 1960 and resulted in severe human poisonings. The second outbreak of methylmercury poisoning from grain consumption occurred in the early 1970s. Imported mercury-treated seed grains arrived after the planting season and were subsequently used as grain to make into flour that was baked into bread. Unlike the long-term exposures in Japan, the epidemic of methylmercury poisoning in Iraq was short in duration, but the magnitude of the exposure was high. Because many of the people exposed to methylmercury in this way lived in small villages in very rural areas (and some were nomads), the number of people exposed to these mercury-contaminated seed grains is not known. The number of people admitted to the hospital with symptoms of poisoning has been estimated to be approximately 6,500, with 459 fatalities reported.

As in the Japanese poisoning epidemics, the signs and symptoms of disease were predominantly those of the nervous system: difficulty with peripheral vision or blindness, sensory disturbances, incoordination, impairment of walking, slurred speech and in some cases, death. Children were affected, as well as adults. Of great concern was the observation that infants, born of mothers who had consumed the methylmercury-contaminated grain (particularly during the second trimester of pregnancy) could show nervous system damage even though the mother was only slightly affected herself.

Toxicokinetics of Mercury

The toxicokinetics (i.e., absorption, distribution, metabolism, and excretion) of mercury is highly dependent on the form of mercury to which a receptor has been exposed.

The absorption of elemental mercury vapor occurs rapidly through the lungs, but it is poorly absorbed from the gastrointestinal tract. Once absorbed, elemental mercury is readily distributed throughout the body; it crosses both placental and blood-brain barriers. The distribution of absorbed elemental mercury is limited primarily by the oxidation of elemental mercury to the mercuric ion as the mercuric ion has a limited ability to cross the placental and blood-brain barriers. Once elemental mercury crosses these barriers and is oxidized to the mercuric ion, return to the general circulation is impeded, and mercury can be retained in brain tissue. Elemental mercury is eliminated from the body via urine, feces, exhaled air, sweat, and saliva. The pattern of excretion changes depending upon the extent the elemental mercury has been oxidized to mercuric mercury.

Absorption of inorganic mercury through the gastrointestinal tract varies with the particular mercuric salt involved; absorption decreases with decreasing solubility. Estimates of the percentage of inorganic mercury that is absorbed vary; as much as 20% may be absorbed. Inorganic mercury has a reduced capacity for penetrating the blood-brain or placental barriers. There is some evidence indicating that mercuric mercury in the body following oral exposures can be reduced to elemental mercury and excreted via exhaled air. Because of the relatively poor absorption of orally administered inorganic mercury, the majority of the ingested dose in humans is excreted through the feces.

Methylmercury is rapidly and extensively absorbed through the gastrointestinal tract. Absorption information following inhalation exposures is limited. This form of mercury is distributed throughout the body and easily penetrates the blood-brain and placental barriers in humans and animals. Methylmercury in the body is considered to be relatively stable and is only slowly demethylated to form mercuric mercury in rats. It is hypothesized that methylmercury metabolism may be related to a latent or silent period observed in epidemiological studies observed as a delay in the onset of specific adverse effects. Methylmercury has a relatively long biological half-life in humans; estimates range from 44 to 80 days. Excretion occurs via the feces, breast milk, and urine.

The most common biological samples analyzed for mercury are blood, urine and scalp hair. The methods most frequently used to determine the mercury levels in these sample types include atomic absorption spectrometry, neutron activation analysis, X-ray fluorescence and gas chromatography.

Some Limitations of the Assessment

In both the Iraqi and Japanese epidemics, the levels of methylmercury consumed were much higher than the levels currently reported in the U.S. food supply. While there are no data to indicate that methylmercury absorption is affected by food type, it must be noted that one of the severe poisoning episodes was through a means not expected to be prevalent in the U.S.; that is, the consumption of contaminated grain.

Health endpoints other than neurotoxicity were evaluated by U.S. EPA using established risk assessment Guidelines. Data for other endpoints than developmental neurotoxicity were limited. Methylmercury has been shown to cause tumors in mice at high doses that produce severe non-cancer toxicity. Low-dose exposures to methylmercury are not likely to cause cancer in humans. Data on effects related to mutation formation (changes in DNA) indicate that methylmercury could increase

What Is A Reference Dose?

A reference dose or RfD is defined in the following way by U.S. EPA: an estimate (with uncertainty spanning perhaps an order of magnitude) of a daily exposure to the human population (including sensitive subgroups) that is likely to be without an appreciable risk of deleterious effects during a lifetime. RfDs are reviewed by Agency scientists for accuracy, appropriate use of risk assessment methodology, appropriate use of data and other scientific issues. When consensus has been reached by the workgroup, information on the RfD is made available to the public through a U.S. EPA database; namely, the Integrated Risk Information System (IRIS).

The RfD is based on the best available data that indicate a "critical effect"; this is generally the first indicator or most subtle indicator of an adverse effect in the species under study. In calculating RfDs U.S. EPA generally uses a no observed adverse effect level (NOAEL). This is found from either inspection or modeling of dose-response data on the critical effect. It is a means of estimating the threshold for effect in the reported study. The NOAEL is most useful when it is from a study in which a determination of the lowest observed adverse effect level (LOAEL) can also be done. The LOAEL is the lowest tested dose at which the critical effect was seen in the species under study.

In calculating the RfD the U.S. EPA divides the NOAEL or LOAEL by a series of uncertainty and modifying factors in order to extrapolate to the general human population. The uncertainty factors (which may be as much as 10 each) are for the following areas: extrapolation of data to sensitive human subpopulations; extrapolation from animal data to conclusions for humans; lack of chronic data; lack of certain other critical data; and use of a LOAEL in the absence of a NOAEL.

The RfD is used for risk assessment judgments dealing with evaluations of general systemic toxicity. It is intended to account for sensitive (but not hypersensitive) members of the human population; the rationale is that if exposure to the RfD is likely to be without appreciable risk for sensitive members of the population, then it is without appreciable risk for all members of the population. The RfD is generally applicable to men and women and to adults, to children and to the aged, unless data support the calculation of separate RfDs for these groups.

The RfD is a quantitative estimate of levels expected to be without effect. Exceedance of the RfD does not mean that risk will be present. Acceptability of uncertain risks is a risk management decision. Risk management decisions may consider the RfD but will take into account exposures, other risk factors and non-risk factors as well.

frequencies of mutation in human eggs and sperm. These data were not sufficient, however, to permit estimation of the amount of methylmercury that would cause a measurable mutagenic effect in a human population.

How Much Methylmercury is Harmful to Humans?

Information on the amount of methylmercury exposure producing particular combinations of signs and symptoms in people has been analyzed to yield what are called quantitative dose-response assessments. Both the Japanese and Iraqi epidemics are important to understanding how methylmercury from food produces neurological disease in humans. In the epidemics in Minamata and Niigata, the exposures were long-term, and the tissues of fish and shellfish were the sources of methylmercury exposure. This establishes with highest scientific confidence that methylmercury in fish can produce human disease. A limitation to these data is that many patients were severely affected. The extent of methylmercury poisoning was so severe that finding subtle indications of disease is difficult. Subtle

indicators of poisoning are important in identifying levels of exposure which will not cause any adverse effects. The U.S. EPA calculates one such measure, called a reference dose or RfD (see the box above).

U.S. EPA has on two occasions published RfDs for methylmercury which have represented the Agency consensus for that time. These are discussed at length in Volume IV, and the uncertainties and limitations are described in Volume VI. At the time of the generation of the Mercury Study Report to Congress, it became apparent that considerable new data on the health effect of methylmercury in humans were emerging. Among these are large studies of fish or fish and marine mammal consuming populations in the Seychelles and Faroes Islands. Smaller scale studies are in progress which describe effects in populations around the U.S. Great Lakes. In addition, there are new evaluations, including novel statistical approaches and application of PBPK models to published work. As the majority of these new data are either not yet published or have not yet been subject to rigorous review, it was decided that it was premature for U.S. EPA to make a change in the methylmercury RfD at this time. An interagency process, with external involvement, will be undertaken for the purpose of review these new data, their evaluations and evaluations of existing data. An outcome of this process will be assessment by U.S.EPA of its RfD for methylmercury to determine if change is warranted.

The current U.S. EPA RfD for methylmercury was based on data on neurologic changes in 81 Iraqi children who had been exposed *in utero*: their mothers had eaten methylmercury-contaminated bread during pregnancy. The data were collected by interviewing the mothers of the children and by clinical examination by pediatric neurologists approximately 30 months after the poisoning episode. The incidence of several endpoints (including late walking, late talking, seizures or delayed mental development, and scores on clinical tests of nervous system function) were mathematically modeled to determine a mercury level in hair (measured in all the mothers in the study) which was associated with no adverse effects. These effects were delays in motor and language development defined by the following:

- Inability to walk two steps without support by 2 years of age;
- inability to respond to simple verbal communication by age 2 years among children with good hearing;
- scores on physical examination by a neurologist that assessed cranial nerve signs, speech, involuntary movements, limb tone, strength, deep tendon reflexes, plantar responses, coordination, dexterity, primitive reflexes, sensation, posture, and ability to sit, stand, walk, and run; and
- assessment of mental development or the presence of seizures based on interviews with the child's mother.

In calculating the mercury level in hair which was associated with no adverse effects, the U.S. EPA chose a benchmark dose (in this instance the lower bound for 10 percent risk of neurological changes) based on modeling of all effects in children. This lower bound was 11 ppm hair concentration for methylmercury. A dose–conversion equation was used to estimate a daily intake of 1.1 µg methylmercury/kg body weight/day that when ingested by a 60 kg individual will maintain a blood concentration of approximately 44 µg/L of blood or a hair concentration of 11 µg mercury/gram hair (11 ppm).

Data on the behavior of mercury in the human body were used to estimate the amount of mercury ingested per day at this no adverse effect level. Due to variability in the way individuals process methylmercury in the body and the lack of data on observed adult male and female reproductive effects, an uncertainty factor of 10 was used to derive the RfD from the benchmark dose. The RfD for methylmercury

was determined to be 1×10^{-4} mg/kg-day; that is a person could consume 0.1 µg methylmercury for every kg of his/her body weight every day for a lifetime without anticipation of risk of adverse effect. The RfD is a daily ingestion level anticipated to be without adverse effect to persons, including sensitive subpopulations, over a lifetime. The RfD may be considered the midpoint in an estimated range of about an order of magnitude. This range reflects variability and uncertainty in the estimate.

The RfD is a risk assessment tool, not a risk management decision. Judgments as to a "safe" dose and exposure represent decisions that involve risk management components.

Limitations and Uncertainties in the Assessment

The range of uncertainty in the RfD and the factors contributing to this range were evaluated in qualitative and quantitative uncertainty analyses. The uncertainty analyses indicated that paresthesia (numbness or tingling) in the hands and feet, and occasionally around the mouth, in adults is not the most reliable endpoint for dose-response assessment because it is subject to the patient's recognition of the effect. Paresthesia in adults is no longer the basis for U.S. EPA's methylmercury RfD as it was in the mid-1980s. There are, however, uncertainties remaining on the current RfD based on developmental effects from methylmercury in children exposed *in utero*. There are difficulties with reliability in recording and classifying events like late walking in children, especially as the data were collected approximately 30 months after the child's birth. It should be noted, however, that the endpoints used represented substantial developmental delays; for example, a child's inability to walk two steps without support at two years of age, inability to talk based on use of two or three meaningful words by 24 months, or presence of generalized convulsive seizures. There is uncertainty in the physiologic factors which were used in estimating the ingested mercury dose. There is also a degree of uncertainty introduced by the size of the study population (81 mother-child pairs). Nevertheless, the RfD for methylmercury is a reasonable estimate based on currently available data.

The RfD is supported by additional studies in children exposed *in utero*. These include investigations among Cree Indians in Canada and New Zealanders consuming large amounts of fish. In these studies the hair concentration of mercury is used to monitor mercury exposure over time. Conclusions by the investigators in their official reports cite developmental delays among the children born of mothers whose hair mercury concentrations during pregnancy were 6 to 18 ppm, consistent with the benchmark dose of 11 ppm.

Currently a number of research studies are underway that further address the question of what exposures to methylmercury in fish are associated with neurological disease. These studies include more subjects than did the Iraqi study, are prospective in design, and utilize endpoints that are anticipated to be more sensitive than the clinical signs and symptoms of methylmercury poisoning observed in Iraq. These studies of fish consumption, rather than poisoning, are conducted in the Seychelles Islands in the Indian Ocean, the Faroe Islands in the North Atlantic Ocean (sponsored by the Department of Health and Human Services), and in the United States; this last study is sponsored by the Agency for Toxic Substances and Disease Registry (ATSDR). Data from these studies, when available, should be useful in decreasing the uncertainty surrounding both the benchmark dose and the RfD. The U.S. FDA has determined that revisions of its action level for mercury concentrations of fish in interstate commerce should wait until the new studies have reduced the level of uncertainty. The availability of results from the above studies will likewise enable U.S. EPA to re-examine and adjust its RfD as needed.

Levels of Methylmercury Exposure Addressed by the U.S. Food and Drug Administration, World Health Organization and State Recommendations

The U.S. EPA RfD is a daily intake level and is a risk assessment tool; the use of the RfD is not limited to fish. The discussion that follows covers risk assessment and risk management activities concerning fish. These consider fish consumption patterns and risk management policy factors. The existing advice and action level of the U.S. FDA is compatible with the U.S. EPA RfD and the assessment information presented in this Mercury Study.

There are numerous local and state warnings in the U.S. to limit intake of fish because of chemical contamination. Warnings are issued because of a number of contaminants. Methylmercury is most often included as one of the contaminants that form the basis for the warning. Often these warnings are issued based on local conditions.

Recommended limits on methylmercury exposure have been expressed in these units: g/kg body weight/day; concentrations of mercury in tissues such as blood, hair, feathers, liver, kidney, brain, etc.; grams of fish per day; number of fish meals per time interval (e.g., per week). Reference values for mercury concentrations (expressed as total mercury) in biological materials commonly used to indicate human exposures to mercury were published by the WHO/IPCS (1990). The mean concentration of mercury in whole blood is approximately 8 g/L, in hair about 2 g/g, and in urine approximately 4 g/L. Wide variation occurs about these values (WHO/IPCS, 1990).

A number of different estimates exist for hair mercury levels that are associated with low risks of neurological endpoints such as paresthesia. These estimates are sensitive to variables such as the half-life of mercury in the body (time to eliminate half the dose of mercury). Half life is usually estimated as an average of 70 days, with extremes of about 35 to just over 200 days reported for different individuals. The half-life of mercury in pregnant women has not been directly measured. The half-life of mercury in women during lactation is shorter, possibly due to excretion of mercury into milk produced during lactation.

Cross-comparison of World Health Organization (WHO) recommendations regarding risk associated with hair mercury concentrations is facilitated by data reported by the WHO on mercury concentrations in 559 samples of human head hair from 32 locations in 13 countries. The WHO report found that mercury concentrations in hair increased with increasing frequency of fish consumption (see Table 3-3).

Table 3-3
WHO Data on Mercury in Hair

| Fish Consumption Frequency | Average Mercury Concentration in Hair (g mercury per g of hair) |
|-----------------------------------|--|
| No unusual mercury exposure | 2 |
| Less than one fish meal per month | 1.4 (range 0.1 to 6.2) |
| Fish meals twice a month | 1.9 (range 0.2 to 9.2) |
| One fish meal a week | 2.5 (range 0.2 to 16.2) |
| One fish meal each day | 11.6 (range 3.6 to 24.0) |

The World Health Organization's International Programme for Chemical Safety (WHO/IPCS) concluded that the general population of adults (males and non-pregnant females) does not face a significant health risk from methylmercury when hair mercury concentrations are under 50 µg mercury/gram hair. In recent evaluations of the Niigata epidemic of Minamata disease, study authors reported lower thresholds with mean values in the range of 25 to approximately 50 µg mercury/gram hair.

Clinical observations in Iraq suggest that women during pregnancy are more sensitive to the effects of methylmercury with fetuses at particularly increased risk. The WHO/IPCS (1990) analyzed the Iraqi data and identified a 30 percent risk to the infant of abnormal neurological signs when maternal hair mercury concentrations were over 70 µg/g. Using an additional statistical analysis, WHO/IPCS estimated a 5 percent risk of neurological disorder in the infant when the maternal hair concentration was 10 to 20 µg mercury/gram of hair. The U.S. EPA RfD is within an order of magnitude of the dose described by WHO.

WHO/IPCS recommended that as a preventive measure, in a subpopulation that consumes large amounts of fish (for example, one serving or 100 grams per day), hair levels for women of child-bearing age should be monitored for methylmercury.

The WHO/IPCS estimated (1990) that a daily methylmercury intake of 0.48 µg mercury/kg body weight will not cause any adverse effects to adults and that a methylmercury intake of 3 to 7 µg/kg body weight/day would result in a <5 percent increase in the incidence of paresthesia in adults. Risk to this extent would be associated with hair mercury concentration of approximately 50 to 125 µg mercury per gram hair. By comparison, the U.S. EPA's reference dose, or the amount of methylmercury any person (including children and pregnant women) can ingest every day without harm is 0.1 µg/kg body weight per day. This was based on a benchmark dose equal to 11 ppm (µg/g) hair. Children are expected to have a higher exposure to methylmercury (on a per kg body weight basis) than do adults.

In 1969, in response to the poisonings in Minamata Bay and Niigata, Japan, the U.S. FDA proposed an administrative guideline of 0.5 ppm for mercury in fish and shellfish moving in interstate commerce. This limit was converted to an action level in 1974 (Federal Register 39, 42738, December 6, 1974) and increased to 1.0 ppm in 1979 (Federal Register 44, 3990, January 19, 1979) in recognition that exposure to mercury was less than originally considered. In 1984, the 1.0 ppm action level was converted from a mercury standard to one based on methylmercury (Federal Register 49, November 19, 1984).

The action level takes into consideration the tolerable daily intake (TDI) for methylmercury, as well as information on seafood consumption and associated exposure to methylmercury. The TDI is the amount of methylmercury that can be consumed daily over a long period of time with a reasonable certainty of no harm. U.S. FDA (and WHO) established a TDI based on a weekly tolerance of 0.3 mg of total mercury per person, of which no more than 0.2 mg should be present as methylmercury. These amounts are equivalent to 5 and 3.3 µg, respectively, per kilogram of body weight. Using the values for methylmercury, this tolerable level would correspond to approximately 230 µg/week for a 70 kg person or 33 µg/person/day. The TDI was calculated from data developed in part by Swedish studies of Japanese individuals poisoned in the episode of Niigata which resulted from the consumption of contaminated fish and shellfish and the consideration of other studies of fish-eating populations.

Based on observations from the poisoning event later in Iraq, U.S. FDA has acknowledged that the fetus may be more sensitive than adults to the effects of mercury (Federal Register 44: 3990, January 19, 1979; Cordle and Tollefson, 1984, U.S. FDA Consumer, September, 1994). In recognition of these concerns, U.S. FDA has provided advice to pregnant women and women of child-bearing age to limit their consumption of fish known to have high levels of mercury (U.S. FDA Consumer, 1994). U.S. FDA believes, however, that given existing patterns of fish consumption, few women (less than 1%) eating such high mercury fish will experience slight reductions in the margin of safety. However, due to the uncertainties associated with the Iraqi study, U.S. FDA has chosen not to use the Iraqi study as a basis for revising its action level. Instead, the U.S. FDA has chosen to wait for findings of prospective studies of fish-eating populations in the Seychelles Islands and in the Faroes Islands.

Characterization of Risk to Human Populations

The characterization of risk to U.S. human populations focuses on exposure to methylmercury. Although methylmercury is found in other media and biota, it accumulates to the highest concentrations in the muscle tissue of fish, particularly fish at the top of the aquatic food chain. As a result, fish ingestion is the dominant exposure pathway. The dominance of this pathway reflects both bioaccumulation of methylmercury in the fish and the efficiency with which methylmercury passes through intestinal walls. The critical elements in estimation of methylmercury exposure from fish are these: the species of fish consumed; the concentration of methylmercury in the fish; the quantity consumed and the frequency of consumption.

There are three ways to assess the risk to populations from methylmercury exposure. The first way used in this analysis was based on predicted increases in methylmercury concentrations in fish due to anthropogenic emissions coupled with predicted exposure to human (and wildlife) populations. This type of analysis has the advantage of predicting the direct impact of anthropogenic emissions on fish concentrations. The second way risk was assessed was by using dietary surveys to identify the amount and type of fish consumed by populations in the U.S. The advantage of this methodology is that a total exposure from fish can be evaluated, even though the contamination may have come from sources other than anthropogenic emissions. The third way to determine whether members of the population are at risk was to consider hair mercury levels as methylmercury exposures for the general populations are reflected by these levels. This type of assessment would be the best measurement of actual mercury exposure because biological samples are utilized. These three methodologies and conclusions regarding the risk characterization are presented below.

Modeled Anthropogenic Emissions and Predicted Fish Methylmercury Levels

The key issue addressed in the risk characterization was whether anthropogenic mercury emissions from U.S. sources have the potential to increase mercury concentrations in freshwater fish such that subsequent consumption of these fish would result in increased risk to the consumer. Due to limitations in the science regarding methylation and bioaccumulation of mercury in marine environments, the analysis did not address the potential impact of these emissions on marine species of fish.

As described in previous sections, this approach used models to evaluate exposures that result from atmospheric mercury emissions from U.S. sources. Exposure to mercury consumption from fish depends on both the mercury concentration in the fish and the amounts of fish consumed. The modeling analysis predicted that some of the mercury emitted from local emission sources deposits on local watersheds and water bodies where a fraction of it is methylated and incorporated into the aquatic food chain. Since mercury emissions are also transported across great distances, the deposition of mercury from distant sources were also considered to contribute to the environmental loading of mercury around a single source. As noted in the discussion of the exposure analysis above, the U.S. EPA concludes that there is a plausible link between anthropogenic emissions and increases in methylmercury concentrations in freshwater fish.

The highest levels of methylmercury in fish (e.g., greater than 1 ppm) were predicted in the trophic level 4 fish; that is, those predator species at the top of the food web. These high predictions generally result from using relatively conservative assumptions. However, the highest predicted fish concentration was 4.9 ppm which is at the high end of the range of measured values in the U.S. Measured values range from less than 0.1 ppm to 8.94 ppm; typical values are between 0.11 and 0.26 ppm.

Local water bodies in proximity (e.g., within 2.5 km) to industrial and combustion sources that emit substantial amounts of divalent mercury from low stacks or at a slow rate appear to be more highly impacted by atmospheric mercury releases. For water bodies located in remote areas, the predicted

concentrations in fish are influenced by the overall proximity to anthropogenic sources, increased soot and ozone concentrations and elevated rainfall.

Given these potential methylmercury concentrations, the issue becomes the fish consumption rate of populations eating fish from these waters. Consumption of fish from these waters was assumed for three types of human populations: an adult with a high fish consumption rate (“high-end consumer”), a child of a high-end consumer and a recreational angler. The consumption and body weights used in the analysis are shown below in Table 3-4.

Table 3-4
Body Weights and Fish Consumption Values Used in Exposure Modeling

| Subpopulation | Assumed Body Weight (kg) | Assumed Local Fish Consumption Rate (g/day) |
|---------------------------|-----------------------------|---|
| Adult High-End Consumer | 70 | 60 |
| Child High-End Consumer | 17 | 20 |
| Adult Recreational Angler | 70 | 30 |

Results of the modeling analysis show that humans consuming fish with predicted mercury concentrations above 1 ppm at the above consumption rates would be ingesting mercury at levels approaching or exceeding the product of 10 times the U.S. EPA’s RfD. It is not possible to predict the degree of risk from exposures above the RfD for methylmercury as the RfD is not a probability estimate. A level of 10 times the RfD is equal to the NOAEL. The NOAEL was the lower bound on a 10 percent effect level; it is not possible to estimate likelihood of risk in a large population exposed to the NOAEL.

A limitation of this modeling analysis is that the size of the population potentially at increased risk cannot be estimated because hypothetical water body locations and exposure scenarios are employed. It is known, however, that there are locations in the U.S. where fish concentrations exceed 1 ppm. For example, the U.S. EPA has found mercury residues in fish at 92 percent of more than 370 surface water bodies tested in the U.S. Mercury levels above 1 ppm were found at 2 percent of the sites surveyed, and above 0.5 ppm at 15 percent of the sites. Figure 3-4 illustrates the geographic location of these sites. In addition, as mentioned previously, a relationship exists between the methylmercury content in fish and lake pH, with higher methylmercury concentrations in fish tissue typically found in more acidic lakes. Although pH is only one parameter that can influence mercury methylation and subsequent bioaccumulation, it is informative to note geographic areas that are both predicted to have elevated mercury deposition rates and include surface waters already impacted by acid deposition. Figure 3-5 displays the regions of the U.S. that meet these criteria.

The potential for a consumer to be at increased risk from fish consumption is modified by at least three important factors. First, many States have issued advisories regarding the consumption of certain species of fish from certain water bodies on account of mercury contamination. These advisories are meant to prevent the public from consuming fish with harmful levels of mercury in them. Thus, exposures to high concentrations are hopefully avoided. (It is known however, that not all anglers heed this advice.)

Figure 3-4

**Distribution of Mercury Concentrations in EPA-Sampled
Fish Tissue Throughout the U.S.**

Figure 3-5
Surface Water with $\text{pH} \leq 5.5$ and Anthropogenic Mercury Deposition

Second, most anglers fish from a variety of water bodies. Several studies indicate that many anglers may travel extended distances to fish. These individuals who consume fish from a variety of locations decrease their chance of exposure to methylmercury at toxicologically significant doses because the extent of mercury contamination can differ significantly between water bodies. Although some areas of the U.S. are known to have fish contaminated with levels above 1 ppm, the national average for freshwater fish (based on the Lowe et al., 1984 data set) is 0.11 ppm, or 0.26 ppm based on data from Bahnick et al., (1985).

Third, some members of the population, even though they consume large quantities of fish, are likely to obtain their fish from both local water bodies and from commercial sources. By eating a variety of fish in the diet, including fish obtained commercially, it is likely that fish with a range of mercury levels are being consumed. A consumer buying fish may be purchasing fish with lower mercury levels than those locally caught. Thus, overall exposure would be reduced. For example, the top ten seafood species that make up 80 percent of the seafood market all have methylmercury levels less than 0.2 ppm. These species are listed in Table 3-5. Note however, that there are some saltwater species, notably shark and swordfish, that do have elevated levels of mercury. These are not frequently consumed species, but their mercury levels are sufficiently high to have potential for increased risk if consumed regularly. Consequently, the FDA advises pregnant women, and women of childbearing age intending to become pregnant, to limit their consumption of shark and swordfish to no more than once a month.

The FDA advises persons other than pregnant women and women of child-bearing age to limit their consumption of fish species with methylmercury levels around 1 ppm to about 7 ounces per week (about 1 serving). For fish with levels averaging 0.5 ppm, regular consumption should be limited to about 14 ounces per week (about two servings). Consumption advice is unnecessary for the top 10 seafood species listed in Table 3-5 as mercury levels are low and few people eat more than the suggested weekly limit of fish (2.2 pounds) for this level of contamination. These consumption advisories are consistent with the findings of the U.S. EPA's modeling analyses described above.

In summary, conclusions that can be drawn from the above discussion are these.

- Emission sources can plausibly be linked with incremental increases in fish methylmercury levels in surface waters in the U.S.
- In some regions of the U.S., fish levels approach or exceed 1 ppm. Increased or continued mercury deposition from anthropogenic emission sources in the U.S. have the potential to increase the mercury concentration in fish above current levels.
- The populations with the highest potential of increased risk are those who routinely and exclusively eat freshwater fish from a single location or region that is known to be impacted by mercury contamination. Consumers are thus urged to heed the advice of state and local health departments concerning local conditions.
- The typical consumer eating fish in moderation from a variety of sources and eating a variety of fish species are not believed to be at increased risk. These consumers are not being advised to limit fish consumption.

Table 3-5
Mercury Concentrations in the Top Ten Types of Fish
Consumed by U.S. Residents

| Fish^a | Mercury Concentration (µg/g, wet weight)^b | Comments |
|-------------------------|---|---|
| Tuna | 0.206 | The mercury content for tuna is the average of the mean concentrations measured in 3 types of tuna: albacore tuna (0.264 µg/g), skipjack tuna (0.136 µg/g) and yellowfin tuna (0.218 µg/g). The U.S. FDA measured the methylmercury concentration in 220 samples of canned tuna in 1991; the average amount of methylmercury measured in these samples was 0.17 g/g and the measured range was <0.1 - 0.75 g/g (Yess, 1993). |
| Shrimp | 0.047 | The mercury content for shrimp is the average of the mean concentrations measured in seven types of shrimp: royal red shrimp (0.074 µg/g), white shrimp (0.054 µg/g), brown shrimp (0.048 µg/g), ocean shrimp (0.053 µg/g), pink shrimp (0.031 µg/g), pink northern shrimp (0.024 µg/g) and Alaska (sidestripe) shrimp (0.042 µg/g). |
| Pollack | 0.15 | The Pesticide and Chemical Contaminant Data Base for U.S. FDA (1991/1992) reports the methylmercury concentration in pollack in commerce as 0.04 g/g. |
| Salmon | 0.035 | The mercury content for salmon is the average of the mean concentrations measured in five types of Salmon: pink (0.019 µg/g), chum (0.030 µg/g), coho (0.038 µg/g), sockeye (0.027 µg/g), and chinook (0.063 µg/g). |
| Cod | 0.121 | The mercury content for cod is the average of the mean concentrations in Atlantic Cod (0.114 µg/g) and the Pacific Cod (0.127 µg/g). |
| Catfish | 0.088 0.16 | The sources of mercury content in catfish are Bahnick et al., 1994 and Lowe et al., 1985. Both data sets were collected from U.S. freshwater sources. The Bahnick data (mean = 0.088) include channel, largemouth, rock, striped and white catfish, and the Lowe data (mean = 0.16) include channel and flathead catfish. It should be noted that neither survey included farm-raised catfish, which is the type of catfish predominantly consumed in the U.S. The mercury content of farm-raised catfish may be significantly different than freshwater sources. The Pesticide and Chemical Contaminant Data Base for U.S. FDA (1991/1992) reports the methylmercury concentration in catfish as 0.02 g/g. |
| Clam | 0.023 | The mercury content for clam is the average of the mean concentrations measured in four types of clam: hard (or quahog) clam (0.034 µg/g), Pacific littleneck clam (0 µg/g), soft clam (0.027 µg/g), and geoduck clam (0.032 µg/g). |

Table 3-5 (continued)
Mercury Concentrations in the Top Ten Types of Fish
Consumed by U.S. Residents

| Fish ^a | Mercury Concentration (µg/g, wet weight) ^b | Comments |
|---------------------|---|---|
| Flatfish (Flounder) | 0.092 | The mercury content for flounder is the average of the mean concentrations measured in nine types of flounder: Gulf (0.147 µg/g), summer (0.127 µg/g), southern (0.078 µg/g), four-spot (0.090 µg/g), windowpane (0.151 µg/g), arrowtooth (0.020 µg/g), witch (0.083 µg/g), yellowtail (0.067 µg/g), and winter (0.066 µg/g). |
| Crab | 0.117 | The mercury content for crab is the average of the mean concentrations measured in five types of crab: blue crab (0.140 µg/g), dungeness crab (0.183 µg/g), king crab (0.070 µg/g), tanner crab (<i>C. opilio</i>) (0.088 µg/g), and tanner crab (<i>C. bairdi</i>) (0.102 µg/g). |
| Scallop | 0.042 | The mercury content for scallop is the average of the mean concentrations measured in four types of scallop: sea (smooth) scallop (0.101 µg/g), Atlantic Bay scallop (0.038 µg/g), calico scallop (0.026 µg/g), and pink scallop (0.004 µg/g). |

^a List of fish types from U.S. FDA (1995).

^b Mercury concentrations sources are described in the comments, refer to Volume III for complete citations.

Human Exposure to Methylmercury Based on Dietary Surveys

The discussion above focused on potential risk to human populations due to consumption of fish having relatively high concentrations of mercury. The analysis of mercury exposure using dietary surveys is aimed at identifying populations that eat much greater amounts of fish than the average consumer. Their potential for increased risk is not necessarily due to elevated concentrations in fish, it is more a function of the amount of fish consumed on a regular, usually daily, basis.

The analysis of the at-risk population eating above average amounts of fish focuses on that part of the population which consumes on average 100 grams or more of fish or shellfish per day (approximately 3.5 ounces). The basis for this focus on persons eating 100 grams or more is a recommendation made by the World Health Organization's International Programme for Chemical Safety (WHO). The WHO's recommendation is that as a preventive measure, in a subpopulation that consumes large amounts of fish (for example, 100 grams per day), hair levels for women of child-bearing age should be monitored for methylmercury.

General U.S. Population. Three groups are potentially at increased risk from methylmercury: pregnant women, women of child-bearing age (i.e., between the ages of 15 and 44) and children ages 14 and younger. Pregnant women are of concern because of the adverse effects of methylmercury on the fetal nervous system. Women of child-bearing age rather than only pregnant women are of concern for two reasons. The first is that methylmercury persists in tissues. Measured half-lives for methylmercury in adults range from about 1 month to 9 months, although half-lives of just over 2 months are usually

observed. Thus, dietary intakes just prior to pregnancy are of concern rather than only methylmercury intakes during pregnancy. The second reason is that women usually do not know they are pregnant until the pregnancy is past many of the critical stages of fetal development.

Children may be at a higher risk of methylmercury exposure than are adults because they appear to have higher exposures on a per kilogram body weight basis, and they may be inherently more sensitive than adults given the developmental state of the nervous system. In the methylmercury poisoning epidemics in Japan and Iraq, children were affected, as well as adults. These effects were not seen only in children exposed to methylmercury *in utero*, but included children exposed through ingesting methylmercury from food. Whether or not children differ from adults in sensitivity to methylmercury neurotoxicity is not known.

The 100 gram per day recommendation by the WHO can be used as a screening analysis to identify populations potentially at increased risk. The significance of the risk is, as mentioned above, is also a function of the methylmercury concentrations of the fish consumed. The U.S. EPA used two types of dietary surveys to identify these populations.

Dietary surveys can be classified into longitudinal or cross-sectional surveys. Cross-sectional data are used to give a "snap shot" in time and are typically used to provide information on the distribution of intakes for groups within the population of interest. Cross-sectional data typically are for 24-hour or 3-day sampling periods and may rely on recall of foods consumed following questioning by a trained interviewer, or may rely on written records of foods consumed. The cross-sectional survey used in the Report was the Continuing Surveys of Individual Food Consumption for the period 1989 to 1991. Typically long-term or longitudinal estimates of intake can be used to reflect patterns for individuals (e.g., dietary histories); or longitudinal estimates of moderate duration (e.g., month-long periods) for individuals or groups. A longitudinal survey used for comparative purposes in this Report is the National Purchase Diary, Inc survey. Additional discussion of these issues are found on Appendix H to Volume III.

The U.S. EPA first evaluated data on fish consumption for a general population in the United States based on the United States Department of Agriculture's Continuing Surveys of Individual Food Consumption for the period 1989 to 1991 (CSFII 89/91). (The survey and analyses are described in detail Appendix H to Volume III and discussed in Volume VI.) The CSFII 89/91 was a cross-sectional survey conducted over a three-year period. Participants kept three-day food consumption diaries which recorded the amount of various foods consumed as well as self-reported body weight. Data were collected over all months of the year and included week days as well as weekends.

During the past decade, reviewers of dietary survey methodology (for example, the Food and Nutrition Board of the National Research Council/National Academy of Sciences; the Life Sciences Research Office of the Federation of American Societies of Experimental Biology) have evaluated various dietary survey techniques with regard to their suitability for estimating exposure to contaminants and intake of nutrients. Having evaluated a number of data sets, the Academy's Subcommittee concluded that 3 days of observation may be more than is required for the derivation of the distribution of usual intakes.

For comparison, the U.S. EPA also used results of a longitudinal food consumption survey; namely, the National Purchase Diary, Inc. data on fish consumption which was conducted between 1973 and 1974 (NPD).

Based on the reported quantities of fish consumed in CSFII 89/91, it was estimated that 2 to 5 percent of the population of women who consume fish eat, on average, 100 grams of fish per day. The CSFII 89/91 does not include the States of Alaska and Hawaii. Because of their substantial coastal areas, fish consumption in these states is likely to be higher than in the continental U.S. Thus, the population

calculated to consume fish in excess of 100 grams per day may be underestimated using this survey. In the one-month sampling period surveys conducted by NPD about 1 percent of the study population consumed over 100 grams of fish per day. Thus, the results of the two surveys are in substantial agreement. Using 1990 census data, the number of women of child-bearing age and of children were calculated with the result multiplied by the percentages above to provide the size of the various populations consuming greater than 100 grams of fish per day. The number of women of child-bearing age who are also pregnant was calculated using the public health statistic that in any given year approximately 9.5 percent of women are pregnant. The results of these analyses are summarized below in Table 3-6. The range represents the results of both dietary surveys.

Table 3-6
Size of the Populations of Concern Consuming 100 Grams or More of Fish Per Day

| Population of Concern | Size of the Population (Represents 1 to 5 percent of total population) |
|------------------------------|---|
| Women of Child-Bearing Age | 547,000 - 886,000 |
| Pregnant Women | 50,000 - 86,000 |
| Children | 503,000 - 665,000 |

The 50,000 estimate is based on the NPD consumption data from the 1970s. Fish consumption has been reported to increase, on average, 26% between 1970 and 1990. Because it is uncertain whether this increase applies to the extremes of the distribution, no attempt was made to adjust the estimate derived from the 1973/74 NPD data set.

Populations Consuming Greater than 100 Grams of Fish Per Day. Populations that consume fish at 100 grams per day or higher are likely to include several segments of the U.S. population. The term "subsistence fishers" has been used to describe various persons who rely on fish as a major source of protein. "Subsistence fishers" are not defined by whether the fish/shellfish are self-caught or purchased. Groups with high fish intake are typically determined by social, economic, ethnic, and geographic characteristics. An additional group of people consumes high levels of fish in response to numerous health-based messages that have promoted the consumption of fish to reduce the likelihood of disease, particularly of the cardiovascular system. Furthermore, there are large numbers of people who simply prefer fish and shellfish as a source of protein. These consumers are represented by these groups: recreational anglers, members of some Native American Tribes, members of ethnic groups who consume higher than typical intakes of fish, persons who preferentially select fish for health-promotion purposes, individuals who prefer the taste of fish, and persons who rely on self-caught fish from local sources because of limited money to buy food. All of these groups may be more reliant on local sources of self-caught fish than is the general population.

Whether or not the dietary surveys of the general population described above adequately represent subpopulations, such as recreational anglers, subsistence fishers, or Native Americans remains a concern. Some groups, for example native populations in Alaska, consume on average, quantities of fish and marine mammals far higher than the overall U.S. population. A review of the published literature on quantities of fish eaten by groups that include anglers, subsistence fishers, and some Native Americans shows that average fish intake is higher for these groups than for the general population. Figure 3-6 illustrates the various consumption rates of these groups.

Estimates of mercury exposure for these consumers can be made based on the following scenario. The average mercury concentration of the mixture of fish/shellfish reported in CSFII 89/91 to be consumed by persons surveyed is 0.134 ppm. As a result, a 100 gram serving would contain 13.4 g mercury per 100 grams. The quantities of fish and shellfish consumed by women of child-bearing age at the 95th, 97.5th and 99th percentile of respondents in CSFII 89/91 are 111, 133 and 175 grams per day, respectively. Analysis of the CSFII 89/91 data also indicated that 33 percent of fish and shellfish come from freshwater and estuarine species. Data for Wisconsin anglers and for members of two tribes of Native Americans living near Puget Sound support the observation that persons who consume freshwater or estuarine fish, also purchase a portion of their fish/shellfish in the marketplace.

Based on the scenario that both locally caught and commercial fish are consumed, consumption of fish from local sources would increase exposure above that which would be expected if only commercial fish were consumed, if the fish from local sources contained more than 0.134 ppm mercury. If local sources are lower in mercury than 0.134 ppm, exposure to local sources could reduce overall mercury exposure. However, if fish high in the aquatic food web are preferentially consumed, mercury exposure would increase. Data from several states indicate that mercury concentrations of locally caught fish are likely to be higher than 0.134 ppm.

Determining actual methylmercury intake for groups that rely on locally caught fish or fish obtained from a limited geographic region or from only a few species of fish requires individual assessment. These particular groups of women and children would benefit from a more specific evaluation. An example of such evaluation is a biological monitoring program based on analyses of hair or blood for mercury.

In summary, the U.S. EPA analysis indicates that the commercial U.S. fish supply is safe for the U.S. population who consume less than 100 grams/day of fish and shellfish, and a wide variety of fish types. Those consumers who eat large quantities of predatory marine species may be at some level of risk from exposure to methylmercury. Consumers of freshwater fish are also, in general, not expected to be at an elevated risk level, unless their sources of fish are contaminated with more than average levels of mercury.

Hair Mercury Measurements

Actual measurements of hair mercury levels would be the best way to assess mercury exposure and risk because mercury exposure is reflected by hair mercury levels. Because fish are the primary exposure pathway for methylmercury there is a broad-based scientific literature describing increases in hair mercury concentrations with increases in fish consumption. Maternal hair mercury concentrations predict mercury concentrations in fetal brain, fetal blood, umbilical cord blood and newborn hair.

The WHO has concluded that the general population of adults (males and non-pregnant females) does not face a significant health risk from methylmercury when hair mercury concentrations are under 50 µg mercury/gram hair. However, in recent evaluations of the Niigata epidemic of

Figure 3-6
Distribution of Fish Consumption Rates of Various Populations

Minamata disease, study authors reported lower thresholds with mean values in the range of 25 to approximately 50 µg mercury/gram hair.

In addition, clinical observations in Iraq suggest that women during pregnancy are more sensitive to the effects of methylmercury with fetuses at particularly increased risk. The WHO analyzed the Iraqi data and identified a 30 percent risk to the infant of abnormal neurological signs when maternal hair mercury concentrations were over 70 µg/g. Using an additional statistical analysis, WHO estimated a 5 percent risk of neurological disorder in the infant when the maternal hair concentration was 10 to 20 µg mercury/gram of hair.

Although data on hair mercury concentrations from a sample representative of the United States population with adequate documentation of quality assurance/quality control do not exist, data from individual studies conducted within the United States are available and are discussed in Volume VI (See Table 5-8 of that volume). These surveys were conducted in widely diverse geographic areas within the United States. The mean hair mercury concentrations identified for subjects in these studies are typically under 1 µg/g or 1 ppm. For a number of the surveys the detection limit was greater than 1 ppm indicating that a substantial number of zero or trace values were included in the mean concentration. The maximum values reported in these individual surveys range from 2.1 to 15.6 ppm. The highest maximum value (15.6 ppm) was reported from a study that specifically focused on persons from the Florida Everglades who consumed wildlife from this area.

Unpublished data (submitted to U.S. EPA by the U.S. Food and Drug Administration) from the early 1980s on a group of United States women of child-bearing age indicate a mean hair mercury concentration of 0.48 ppm for the overall sample of 1,431 women and a mean hair mercury concentration of 0.52 ppm for the 1,009 women who reported consuming some seafood (statistical analyses of variability in these data [e.g., error estimates] were not included in the data provided to U.S. EPA). These mercury concentrations correspond to hair mercury concentrations associated with fish consumption at the level of less than one meal per month to one meal per week based on data shown in Table 3-3. The magnitude of the increase in hair mercury concentration shown in these unpublished data for those women reporting seafood consumption differs from the patterns observed in most surveys identified in the literature on hair mercury concentrations. This difference between these unpublished data and most literature underscores the need for survey data on hair or blood mercury concentrations from a representative sample of the United States population to estimate body burden of mercury in the general United States population. Until appropriate survey data for the general United States population exist, the overall pattern of hair mercury concentrations for the United States remains unclear. For adequate prediction of methylmercury exposure for the general United States population, the data should be obtained from subjects who are chosen based on a sampling strategy that can be extrapolated to the United States population, and must include appropriate quality assurance/quality control procedures.

Summary of the Risk Characterization

Individuals who consume more than 100 grams (about 3.5 ounces) of fish per day on average may ingest methylmercury in quantities which could pose health effects. Groups such as some Native Americans or subsistence fishermen do consume fish in these large quantities for cultural or economic reasons. The U.S. EPA estimates that between 1 and 5 percent of the U.S. population eats fish in these quantities. The actual risk of exposure depends on the methylmercury concentration of the fish being consumed. These individuals should avoid frequent consumption of fish species which have relatively high methyl mercury concentrations. Pregnant women, women of child-bearing age and children consuming above 100 grams (about 3.5 ounces) of fish per day are the populations of most concern.

Fish which have relatively high concentrations of methylmercury have been identified by the FDA to include shark and swordfish. Fish consumers should also follow advice of States and local authorities regarding fish caught in fresh water bodies for which mercury consumption advisories have been issued.

Limitations of the Assessment

The primary purpose of the Mercury Study Report to Congress was to assess the impact of U.S. anthropogenic emissions on mercury exposure to humans and wildlife. The size of some populations of concern have been estimated; namely women of child-bearing age and children who eat fish. In the general population, people typically obtain their fish from many sources. The question on whether or not the impact of mercury from anthropogenic ambient emissions can be proportioned to the overall impact of methylmercury on wildlife is a much more difficult issue.

As with environmental monitoring data, information on body burden of mercury in populations of concern (blood and/or hair mercury concentrations) are not available for the general U.S. population. Data on higher-risk groups are currently too limited to discern a pattern more predictive of methylmercury exposure than information on quantities of fish consumed. The selenium content of certain foods has been suggestive as a basis for modifying estimates of the quantities of methylmercury that produce adverse effects. Currently, data on this mercury/selenium association form an inadequate basis to modify quantitative estimates of human response to a particular exposure to mercury.

Available data for human health risk assessment have limitations as described in the Report and in this summary. Studies of human fish-consuming populations in the Seychelles and Faroes Islands address some of these limitations; they are expected to be published within a year of release of this Report. Additional studies on U.S. populations who consume fish from the Great Lakes are in progress. Public health agencies of the U.S. government as well as the U.S. EPA will evaluate these new data when they are available.

The benchmark dose methodology used in estimating the RfD required that data be clustered into dose groups. Most data on neurologically based developmental endpoints are continuous; that is, not assigned to dose groups. For example, scoring on scales of IQ involves points rather than a "yes/no" type of categorization. Measurements on the degree of constriction of the visual field involve a scaling rather than a "constricted/unconstricted" type of variable. Although arbitrary scales can be constructed, these groupings have generally not been done in current systems. Use of alternative dose groupings (as described in Volume IV) had no significant effect on calculated benchmark doses. An additional difficulty occurs in estimation of benchmark dose for multiple endpoints that have been measured. Further research on appropriate methods for mathematical modeling is needed. For some situations such information is known, but for methylmercury exposure and multiple endpoints assessing the same system (i.e., developmentally sensitive neurological, neuromotor and neuropsychological effects) the time-course/dose-response of such changes have not been clearly established. Development of the mathematical models needs to be accompanied by understanding the physiological/pathological processes of methylmercury intoxication.

How Much Methylmercury Exposure is Harmful to Wildlife and What Are the Effects?

Massive poisonings of birds and wildlife from methylmercury-treated seed grains were identified during the decades preceding the 1970s. These findings resulted in substantial limitation on use of methylmercury-treated seed grains. However, methylmercury contamination of the aquatic foodchain from many sources continues to adversely affect wildlife and domestic mammals and wild birds. In Minamata, Japan from about 1950–1952 (prior to recognition of human poisonings) severe difficulties with flying and other grossly abnormal behavior was observed among birds. Signs of neurological disease including convulsions, fits, highly erratic movements (mad running, sudden jumping, bumping into objects) were observed among domestic animals, especially cats that consumed seafood.

Generally the place of wildlife in the aquatic foodchain of the ecosystem and their feeding habits determine the degree to which the species is exposed to methylmercury. Fish-eating (piscivorous) animals and those which prey on other fish-eaters accumulate more mercury than if they consumed food from terrestrial food chains. In a study of fur-bearing animals in Wisconsin, the species with the highest tissue levels of mercury were otter and mink, which are top mammalian predators in the aquatic food chain. Top avian predators of aquatic food chains include raptors such as the osprey and bald eagle. Smaller birds feeding at lower levels in the aquatic food chains also may be exposed to substantial amounts of mercury because of their high food consumption rate (consumption/day/gram of body weight) relative to larger birds.

Laboratory studies under controlled conditions can be used to assess the effects of methylmercury from fish on mink, otter and several avian species. Effects can occur at a dose of 0.25 µg/g bw/day or 1.1 µg/g methylmercury in diet. Death may occur in species at 0.1–0.5 µg/g body weight/day or 1.0–5.0 µg/g in the diet. Smaller animals (for example, minks, monkeys) are generally more susceptible to mercury poisoning than are larger animals (for example, mule deer, harp seals). Smaller mammals eat more per unit body weight than larger mammals. Thus, smaller mammals may be exposed to larger amounts of methylmercury on a body weight basis.

Whole body residues of mercury in acutely poisoned birds usually exceed 20 µg/g fresh weight. Although sublethal effects include a number of different organ systems, reproductive effects are the primary concern. These occur at concentrations far lower than those that cause overt toxicity.

The broad ecosystem effects of mercury are not completely understood. No applicable studies of the effects of mercury on intact ecosystems were found. Consequently, characterization of risk for non-human species did not attempt to quantify effects of mercury on ecosystems, communities, or species diversity. The characterization focused on quantities of mercury that adversely affects the health of sensitive subpopulations of wildlife species and on the co-location of these populations with areas of elevated mercury exposure secondary to ambient, anthropogenic emissions of methylmercury. To this end wildlife criteria (WC) were calculated for three piscivorous birds and two mammals (see Table 3-7). The WC is a mercury level in water which is expected to be without harm for the species. The WC considers the bioaccumulation of mercury in the large and small fish eaten by the mammals or birds. A bioaccumulation factor (BAF) was used in the WC calculation; the BAF was based on data on mercury in fish and the water from which they were taken. The effects data for mammals were from a short-term study of neuro-toxicity in mink. The data for fish-eating birds were from a 3-generation study in mallard ducks.

Table 3-7
Wildlife Criteria for Mercury

| Organism | Wildlife Criterion (pg/L) |
|-------------|---------------------------|
| Mink | 415 |
| River otter | 278 |
| Kingfisher | 193 |
| Osprey | 483 |
| Bald eagle | 538 |

The evaluation of data and calculation of WC in this Report was done in accordance with the methods and assessments published in the Final Water Quality Guidance for the Great Lakes System: Final Rule. Availability of additional data led to differences in calculated values of the WC in this Report and those published in the final rule. Differences were the result of three factors. The Report uses more recent data to derive BAF. Second, the final rule appropriately used some region-specific assumptions that were not used in the nationwide assessment in the Report; for example, consumption of herring gulls by eagles. Finally different endpoints were used for the evaluation of mammals because the purposes of the assessments in the Report and final rule were different. In the final rule, a risk-management decision was made to base the wildlife criterion on endpoints likely to influence whole populations (mortality, growth). In this Report a more sensitive endpoint was selected with the goal of assessing the full range of effects of mercury. The difference in the results reflects the amount of discretion allowed under Agency Risk Assessment Guidelines.

In restricted wildlife populations, effects of mercury originating from point sources have been conclusively demonstrated and provide a residue basis for evaluation of mercury levels in animal tissue as an indicator of risk to other populations. Although clear causal links have not been established, mercury originating from airborne deposition may be a contributing factor to population effects on bald eagles, river otters and mink. There is evidence to support the possibility of toxic effects on the common loon and the Florida panther.

Derivation of a WC to protect the Florida panther is complicated by the possibility that prey items (e.g., raccoon) accumulate mercury to an even greater extent than the fish represented by trophic level 4. Other prey (e.g., deer) probably contain relatively lower levels of mercury. Calculation of a WC protective of the panther, therefore, requires collection of additional information on the diet of this species and mercury residues in that diet. Existing data are insufficient to support such an analysis. A chronic NOAEL for domestic cats was reported to be 20 g/kg/d. This is close to that of 5.5 g/kg/d estimated for mink (that is, the subchronic NOAEL of 55 g/kg/d divided by a UF_s of 10). Cats (and presumably larger felines) do not, therefore, appear to be uniquely sensitive or insensitive to the toxic effects of mercury.

Methylmercury (as described in Volumes IV and V of this Report) has deleterious effects on the chordate nervous system. The human health endpoint of concern is developmental neurotoxicity. The health endpoints of concern for the avian wildlife species are reproductive and behavioral deficits and for the mammalian quadrupeds are neurological effects. Assuming that the effects are of similar concern for the well-being of individuals within a species, the NOAELs, LOAELs and the human and wildlife WCs for these health endpoints can then be compared across species.

The human benchmark dose of 11 ppm mercury in hair was considered operationally equivalent to a NOAEL in the derivation of the methylmercury RfD. A LOAEL of 52.5 ppm mercury in hair was estimated for purposes of this risk characterization from inspection of data in Table 5-4 of Volume VI. The NOAEL of 11 ppm mercury in hair and the LOAEL of 52.5 ppm mercury in hair correspond to ingestion levels of 1 g/kg-day and 5.3 g/kg-day, respectively; these dose conversions were made by applying the methods for converting hair mercury concentrations to ingestion levels used in the derivation of the RfD in Volume IV of this Report.

The avian RfD was based on the data from a series of studies by Heinz and collaborators on mallard ducks. A NOAEL could not be identified. The estimated LOAEL, based on reproductive and behavioral effects, was 64 g/kg bw/day. The mammalian RfD was based on the data from a series of studies by Wobeser and collaborators done on ranch mink. A NOAEL of 55 g/kg bw/day was estimated from these studies. The estimated LOAEL, based on damage to the nervous system and liver, was 180 g/kg bw/day.

Based on the data developed for the health assessment, the human LOAEL and RfD are orders of magnitude lower than the corresponding LOAELs and RfD of the other animals (Table 5-5). There is a great deal of uncertainty in this comparison. It must be noted that the effects in humans are based on the RfD definition of a critical effect; that is the most sensitive reported adverse effect or indicator of adverse effect. The effects reported for mammals (i.e., neurologic damage in the mink) and birds (i.e., reproductive effects in mallards) would be considered frank effects in the human RfD methodology. The observations in laboratory animals indicate that it would be reasonable to expect more subtle and less damaging effects of methylmercury to occur at lower doses than the wildlife LOAEL and NOAEL.

The information assessed in this Report suggests that ecosystems most at risk from airborne releases of mercury exhibit one or more of the following characteristics:

- They are located in areas where atmospheric deposition of mercury is high;
- they include surface waters already impacted by acid deposition;
- they possess characteristics other than low pH that result in high levels of bioaccumulation; and/or
- they include sensitive species.

Areas which meet the first two criteria are delineated in Figure 3-5. The adverse effects of methylmercury on wildlife have been described and quantified. For wildlife the importance of site-specific effects of mercury exposure are anticipated to be greater than for humans in the general population because wildlife obtain their fish from a much more limited geographic area than do people.

Limitations of the Assessment

There is uncertainty and variability associated with each WC. These include lack of long-term studies for mammals, lack of a no adverse effects level for birds, and extrapolation from one species to another. It is not known if the species selected for WC development are the most sensitive or appropriate species; also, it is not known if protecting individual animals or species will guarantee protection of their ecosystem from harmful effects of mercury. There are uncertainties and expected variability in the BAF; it was the subject of a quantitative uncertainty analysis.

For wildlife risk assessment, as for humans, mercury toxicity among wildlife involves neurological effects. Available toxicology data from laboratory-based studies of wildlife exposed to methylmercury have measured only gross clinical signs and symptoms of disease and death or pathological changes accompanying these clinically evident changes. Physiologically based evaluation of wildlife has not been done. The importance of more subtle endpoints of neurological function is anticipated to be relevant to such practical questions as the ability of visual hunters such as the loon to find food.

The risk assessment for wildlife made the assumption that the primary source of mercury exposure to the selected species was contaminated fish. Since mercury bioaccumulation is largely through aquatic ecosystems, it is reasonable to focus attention on wildlife species whose feeding habits are tied to these systems. Existing data permit a general treatment of mercury exposure and effects on such populations. For some species, such as the kingfisher and river otter, it can be reasonably assumed that fish always comprise a high percentage of the diet. For others, such as the eagle and mink, considerable variations in diet are likely to exist. Still others, such as the Florida panther, consume prey (such as the raccoon) which consume variable amounts of aquatic biota, but which in South Florida are closely linked to the aquatic food chain. A more accurate characterization of the risk posed by mercury to a specific group of animals occupying a given location will depend on the collection of necessary supporting information: food habits, migratory behavior, breeding biology, and mercury residues in preferred prey items.

To improve the characterization of risk, research needs highlighted in the preceding sections should be addressed. Additional work to decrease uncertainty should be directed toward the exposure assessment. Validated local and regional atmospheric fate and transport models are needed. This should utilize long-term national monitoring networks. Data to improve understanding of movement of mercury through environmental media are also needed. The bioaccumulation factors are major sources of uncertainty. This uncertainty will be decreased by improved data to use in the parameters of the bioaccumulation factor equations and by increased understanding of mercury biogeochemistry in water bodies.

4. MANAGEMENT ALTERNATIVES

Possible Control Strategies

Effective control of mercury emissions may require a mix of strategies including pollution prevention, materials separation and conventional regulatory strategies to control mercury emissions at the stack. Pollution prevention would be suitable for those processes or industries where a mercury substitute is demonstrated and available (e.g., mercury cell chlor-alkali plants). Material separation is an appropriate approach for processes where mercury-containing products are disposed of by incineration, or where mercury can be reduced in the fuel prior to the fuel being combusted (e.g., medical waste incineration). The third approach, conventional regulatory strategies, may be applicable when mercury is emitted to the environment as a result of trace contamination in fossil fuel or other essential feedstock in an industrial process (e.g., smelting). Other non-traditional approaches such as emissions trading or application of a use tax, or other market-based approaches may also prove feasible for mercury control. In addition, emissions control is only one possible means for risk control; reduced human exposure, for example through the use of fish advisories, is another alternative that would need to be explored when selecting among strategies for reducing risks to human health (though not to ecosystems).

The analyses of control technologies and costs presented in this Report are not intended to replace a thorough regulatory analysis, as would be performed for a rulemaking. The information presented is intended to present the range of available options and provide a relative sense of the extent of mercury reductions achievable and the general magnitude of the cost of such reductions.

The three major types of control techniques reviewed are:

- Pollution prevention measures, including product substitution and process modification;
- Materials separation; and
- Flue gas treatment technologies.

Table 4-1 summarizes mercury control techniques for selected source categories.

Pollution Prevention Measures

One possible means of achieving reductions in mercury emissions is through the use of pollution prevention or source reduction. Such approaches to achieving reductions involve changes in processes or inputs to reduce or eliminate emissions of mercury from a particular product or process. They could include, for example, the replacement of mercury with an appropriate substitute or the use of low-mercury content inputs.

In considering opportunities for pollution prevention or source reduction it is important to consider both the potential reductions achievable and the costs of these options. Any consideration of the potential reductions should examine whether (and the extent to which) emission reductions from the particular sources in question will yield reductions in risk to public health and the environment. It is also essential to understand the costs associated with implementing a pollution prevention measure, including any changes in the quality of the end product.

Table 4-1
Summary of Mercury Control Techniques for Selected Source Types

| Mercury Control Technique | Applicable Source Type | Estimated Mercury Removal Efficiency | Cross-Media Impacts?^a | Other Pollutants Controlled | Comments |
|--|----------------------------------|---|---|---|--|
| Product substitution (e.g., batteries, fluorescent lights) | MWCs, MWIs | Variable, depending on the extent of substitution | Yes | Could include other components of mercury-containing batteries, fluorescent lights and other products | <ul style="list-style-type: none"> • Product substitution has reduced the use of mercury in household batteries • Use of mercury-containing fluorescent lights has increased because of their energy efficiency, but lower mercury content is being achieved • The impact of product substitution to other areas depends on specific circumstances, including technical and economic feasibility |
| Process modification | Mercury cell chlor-alkali plants | 100% | Yes | None directly | <ul style="list-style-type: none"> • In 1994, about one-half of the chlor-alkali plants used mercury-free processes • Because the membrane cell process has lower electricity demands than the mercury cell process, plant conversion results in an energy savings • Additional savings presumably also result by avoiding costs of recycling or disposing of mercuric wastes |
| Materials separation | MWCs and MWIs | Variable, depending on the extent of separation | Yes | Could include other components of mercury-containing wastes burned in MWCs or MWIs | <ul style="list-style-type: none"> • Separation of low-volume materials containing high mercury concentrations (e.g., batteries, fluorescent lights, thermostats and other electrical items) can reduce mercury input to a combustor without removing energy content of the waste stream • Household battery separation has been implemented by several communities; program efficiency ranges from 3 to 25 percent • Pilot studies conducted at hospitals have been successful |

Table 4-1 (continued)
Summary of Mercury Control Techniques for Selected Source Types

| Mercury Control Technique | Applicable Source Type | Estimated Mercury Removal Efficiency | Cross-Media Impacts?^a | Other Pollutants Controlled | Comments |
|---|---|--|---|--|---|
| Carbon filter beds | MWCs, utility boilers, industrial boilers | 99% | Yes | Residual organic compounds, other heavy metals, SO ₂ , acid gases | <ul style="list-style-type: none"> • Currently applied to five full-scale power plants in Germany, and planned to be installed on five hazardous waste incinerators in Europe • Technically feasible to other sources, such as MWIs or smelters, but has not been applied • Potential negative effects associated with the disposal of spent carbon and the potential for fires in the bed |
| Wet scrubbing (single-staged polishing scrubbers) | MWCs, MWIs, boilers | Can be >90% for water-soluble species; limited for elemental mercury | Yes | Acid gases, metals, particulate matter, dioxins, furans | <ul style="list-style-type: none"> • Applied to one MWI in the U.S. • Have not been applied to MWCs or boilers in the U.S., although they have been used at MWCs in Europe • Requires treatment of wastewater prior to disposal • May form more toxic, lesser-chlorinated dioxin and furan congeners |
| Depleted brine scrubbing | Chlor-alkali plants | 98% | Yes | None | <ul style="list-style-type: none"> • Very little information is available on this technique |
| Treated activated carbon adsorption | Chlor-alkali plants | 90% | Yes | Residual organic compounds, other heavy metals, SO ₂ , acid gases | <ul style="list-style-type: none"> • Very little information is available on this technique • In 1984, carbon bed systems were in use at 8 of the 20 chlor-alkali plants in operation in the U.S. at that time |

Table 4-1 (continued)
Summary of Mercury Control Techniques for Selected Source Types

| Mercury Control Technique | Applicable Source Type | Estimated Mercury Removal Efficiency | Cross-Media Impacts?^a | Other Pollutants Controlled | Comments |
|----------------------------------|--|---|---|--|---|
| Selenium filters | Primary copper smelters, primary lead smelters, and (more limited) MWCs, crematories, power plants | 90% | Yes | Particulate matter, acid gases | <ul style="list-style-type: none"> • Factors that influence performance include inlet mercury concentrations, flue gas temperature and flue gas dust content • Four known applications at smelters as well as a MWC and a crematory in Sweden; known installation at a German power plant; potentially applicable to MWIs • Spent filter containing selenium and mercury must be landfilled after use • More information needed on the possibility of selenium being emitted from the filter itself |
| Activated carbon injection | MWCs, MWIs, utility boilers | 50-90+% | Yes | Chlorinated dioxins and furans, potentially other semi-volatile organics | <ul style="list-style-type: none"> • Activated carbon injection efficiencies reported for utility boilers are based on pilot-scale data and as such have a high degree of uncertainty • Factors that influence performance include flue gas temperature, amount of activated carbon injected, type of particulate matter collector, concentration and species of mercury in flue gas and type of carbon used • Addition of carbon could have significant impact on amount of particulate matter requiring disposal from utility boilers, but not from MWCs or MWIs |

^a For the purpose of this table, cross-media impacts refer to the potential to transfer and release mercury to media other than air, such as soil, ground water, and surface water. For example, carbon filter beds and wet scrubbers remove mercury from air emissions but result in the generation and disposal of mercury-containing solid and liquid wastes, respectively.

Materials Separation

Removing mercury-containing products such as batteries, fluorescent lights and thermostats from the waste stream can reduce the mercury input to waste combustors without lowering the energy content of the waste stream. The mercury removal efficiency would vary, however, depending on the extent of the separation. Many materials in wastes contain mercury. Materials that comprise a large portion of the waste stream, such as paper, plastic, dirt and grit and yard waste, contain very low concentrations of mercury. Therefore, obtaining appreciable mercury reduction from separation of these types of materials would require separating a large fraction of the total waste stream. Separating these materials would counter the intended purpose of the combustion process, which is to disinfect and reduce the volume of waste materials.

Other materials contain higher concentrations of mercury, but make up only a very small portion (less than 1 percent) of the waste stream. These materials include mercuric oxide batteries, fluorescent lights, thermostats and other electrical items. Separation of such materials can reduce mercury input to a combustor without removing any of the energy content of the waste stream. To evaluate a materials separation program, the feasibility and costs of separating a particular material should be compared with the mercury emission reduction achieved. Furthermore, the current and future mercury reduction achieved by separating a certain material should be considered since the mercury contribution of some materials such as household batteries has already declined considerably.

Coal cleaning is another option for removing mercury from the fuel prior to combustion. In some states, certain kinds of coal are commonly cleaned to increase its quality and heating value. Approximately 77 percent of the eastern and midwestern bituminous coal shipments are cleaned in order to meet customer specifications for heating value, ash content and sulfur content.

There are many types of cleaning processes, all based on the principle that coal is lighter than the pyritic sulfur, rock, clay, or other ash-producing impurities that are mixed or embedded in it. Mechanical devices using pulsating water or air currents can physically stratify and remove impurities. Centrifugal force is sometimes combined with water and air currents to aid in further separation of coal from impurities. Another method is dense media washing, which uses heavy liquid solutions usually consisting of magnetite (finely ground particles of iron oxide) to separate coal from impurities. Smaller sized coal is sometimes cleaned using froth flotation. This technique differs from the others because it focuses less on gravity and more on chemical separation.

Some of the mercury contained in coal may be removed by coal cleaning processes. Volume II of this report (Inventory of Anthropogenic Mercury Emissions in the United States) presents available data on the mercury concentrations in raw coal, cleaned coal and the percent reduction achieved by cleaning. These data, which cover a number of different coal seams in four states (Illinois, Pennsylvania, Kentucky and Alabama), indicate that mercury reductions range from 0 to 64 percent, with an overall average reduction of 21 percent. This variation may be explained by several factors, including different cleaning techniques, different mercury concentrations in the raw coal and different mercury analytical techniques. It is expected that significantly higher mercury reductions can be achieved with the application of emerging coal preparation processes. These include selective agglomeration and advanced column flotation. Bench-scale testing is also being carried out to investigate the use of naturally-occurring microbes to reduce the mercury and other trace elements from coal.

Any reduction in mercury content achieved by coal cleaning results in a direct decrease in mercury emissions from boilers firing cleaned coals. The mercury removed by cleaning processes is transferred to coal-cleaning wastes, which are commonly in the form of slurries. No data are available to assess the emissions of mercury from coal-cleaning slurries.

Flue Gas Treatment Technologies

With the exception of mercury, most metals have sufficiently low vapor pressures at typical air pollution control device operating temperatures that condensation onto particulate matter is possible. Mercury, on the other hand, has a high vapor pressure at typical control device operating temperatures, and collection by particulate matter control devices is highly variable. Factors that enhance mercury control are low temperature in the control device system (less than 150 °Celsius [°C] [300 to 400 °Fahrenheit (°F)]), the presence of an effective mercury sorbent and a method to collect the sorbent. In general, high levels of carbon in the fly ash enhance mercury sorption onto particulate matter which is subsequently removed by the particulate matter control device. Additionally, the presence of hydrogen chloride (HCl) in the flue gas stream can result in the formation of mercuric chloride (HgCl₂), which is readily adsorbed onto carbon-containing particulate matter. Conversely, sulfur dioxide (SO₂) in flue gas can act as a reducing agent to convert oxidized mercury to elemental mercury, which is more difficult to collect.

Add-on controls to reduce mercury emissions are described in detail in Volume VII of this report, including information on commercial status, performance, applicability to the specified mercury emission sources, and secondary impacts and benefits. The controls described are:

- Carbon filter beds;
- Wet scrubbing;
- Depleted brine scrubbing;
- Treated activated carbon adsorption;
- Selenium filters; and
- Activated carbon injection.

The most important conclusions from the assessment of flue gas treatment technologies include:

- Conversion of mercury cell chlor-alkali plants to a mercury-free process is technically feasible and has been previously demonstrated.
- Control technologies designed for control of pollutants other than mercury (e.g., acid gases and particulate matter) vary in their mercury-removal capability, but in general achieve reductions no greater than 50 percent.
- Selenium filters are a demonstrated technology in Sweden for control of mercury emissions from lead smelters. Carbon filter beds have been used successfully in Germany for mercury control on utility boilers and MWCs. These technologies have not been demonstrated in the U.S for any of these source types.
- Injection of activated carbon into the flue gas of MWCs and MWIs can achieve mercury reductions of at least 85 percent. The addition of activated carbon to the flue gas of these source types would not have a significant impact on the amount of particulate matter requiring disposal.
- No full-scale demonstrations of mercury controls have been conducted in the U.S. for utility boilers. Based on limited pilot-scale testing, activated carbon injection provides variable control of mercury for utility boilers (e.g., the same technology might capture 20 percent of the mercury at one plant and 80 percent at another). The most important factors affecting mercury control on utility boilers include the flue gas volume, flue gas temperature and chloride content, the mercury concentration and chemical form of mercury being emitted.

- The chemical species of mercury emitted from utility boilers vary significantly from one plant to another. Removal effectiveness depends on the species of mercury present. To date, no single control technology has been identified that removes all forms of mercury.
- The addition of activated carbon to utility flue gas for mercury control would significantly increase the amount of particulate matter requiring disposal.

A number of research needs were also identified in the area of control technologies. These are included in Section 5 of this volume.

Cost of Controls

The overall approach for assessing the cost of flue gas treatment technologies was to select a subset of source categories on the basis of either their source category emissions in the aggregate or their potential to be significant point sources of emissions. Consideration was also given to whether a particular source category was a feasible candidate for application of a control technology-based standard under section 112 of the CAA. This narrowed the analyses to six source categories: MWCs, MWIs chlor-alkali plants, utility boilers, and primary lead and copper smelters.

In addition to determining the cost effectiveness of applying mercury control technology, a financial analysis was performed to evaluate the affordability of mercury control (in terms of potential price increases or impacts on financial impact) for the selected source categories. This analysis is presented in Volume VII of this report.

Table 4-2 presents the six source categories for which a control technology and cost analysis was performed. The table presents the number of facilities in each category and the percent contribution of each to the national inventory. Potential national mercury reductions, potential national control costs and cost-effectiveness estimates are also presented. These estimates are based on the assumption that all plants within a source category will achieve the same reductions and incur the same costs as the model plants used in the analysis. Because this assumption would not be applicable in all circumstances, the estimates of potential reductions and costs should be used only for relative comparisons among the source categories to give an initial indication as to where mercury controls could provide the most emission reduction for the least cost.

The cost of mercury control incurred by any specific facility may be underestimated by the cost analysis presented in this Report because of variability inherent in the assumptions that were made in the analyses. These include the efficiency of the various control techniques for reducing mercury, the amount of mercury in the flue gas stream and other site-specific factors such as down-time and labor costs. In addition, costs for monitoring and record keeping were not included in the cost analyses. On the other hand, the costs represent retrofit application of controls. Installation of controls at new facilities can be significantly less expensive than retrofitting an existing facility.

Table 4-2
Potential Mercury Emission Reductions and Costs for Selected Source Categories

| Mercury Source Category | Number of Facilities | % of U.S. Mercury Emission Inventory | Mercury Control Techniques | Potential National Reductions ^b | Potential National Annual Costs ^c | Cost-Effectiveness (\$/lb of mercury removed) ^d |
|--|------------------------|--------------------------------------|--|---|--|--|
| Municipal waste combustors | 149 | 23 | Material separation Product substitution <u>Activated carbon injection</u> Carbon filter beds Polishing wet scrubber | 50 tons (90% reduction) | \$56 million | \$211-870 |
| Medical waste incinerators | ~3,700 | 27 | Material separation <u>Activated carbon injection</u> Polishing wet scrubber | 60 tons (90% reduction) | \$24 million | \$228-955 |
| Coal-fired utility boilers | 426 (1,043 boilers) | 21 | Fuel switching Advanced coal cleaning Carbon filter beds <u>Activated carbon injection</u> | 24-44 tons (50-90% reduction) ^e | \$2.9 billion | \$5,240-28,000 |
| Chlor-alkali plants using the mercury cell process | 14 | 2.7 | <u>Process modification</u> Depleted brine scrubbing Treated activated carbon adsorption | 6.5 tons (100% reduction) | \$70 million | \$4,590 |
| Primary copper smelters | 8 | 0.3 ^a | <u>Selenium filters</u> | >0.7 tons (90% reduction) | \$7.7 million | \$497 |
| Primary lead smelters | 3 | 3.7 | <u>Selenium filters</u> | 8 tons (90% reduction) | \$0.8 million | \$1,061 |
| Total | ~4,900 | 78 | | | ~\$3 billion | |

NOTE: The underlined mercury control techniques are the techniques on which potential national reductions and potential national annual costs are based.

^a Reflects one smelter only; a national estimate would be higher.

^b Estimated reductions assuming every facility could achieve the reduction listed.

^c Potential national costs are estimates only and assume all facilities would incur the same costs as the model plants used in the analysis.

^d Where cost-effectiveness values are presented as a range, the values reflect the range across facilities of different sizes.

^e The range in potential national reductions reflects the variable efficiency of activated carbon injection to control mercury emissions from coal-fired utility boilers. Activated carbon injection has not been demonstrated for a full-scale utility boiler application. Control costs are based on the installation of spray cooler, fabric filter and carbon injection systems.

The estimates of cost for mercury reduction also do not illustrate two important considerations. One is that all of the cost of control is attributed to mercury removal. As described previously in this Report, many of these controls achieve reductions of other pollutants as well. The benefits of these additional reductions should also be considered. Second, the technologies available for mercury control represent relatively new applications of these technologies. Thus, it is possible that new or emerging control technologies will improve the cost-effectiveness.

Clean Air Act Provisions Applicable to Mercury Control

Mercury is a priority pollutant across numerous U.S. EPA programs including air, water, hazardous waste and pollution prevention. The focus of this chapter is the statutory authority under the CAA that could be used to control mercury emission sources. A brief summary of these authorities is provided below.

Section 112(a) Lesser Quantity Emission Rates

The U.S. EPA Administrator has the discretion to redefine major sources by setting an emissions cutoff lower than the 10 tons per year emission rate level for a single pollutant or 25 tons per year emission rate for a mixture of pollutants. This is referred to as a lesser quantity emission rate (LQER). The CAA states that LQERs are pollutant-specific and should be based on public health or environmental effects.

The major implications of setting an LQER are that all the requirements for a major source, including setting maximum achievable control technology (MACT) standards, mandatory residual risk analyses, calculation of the MACT floor, modification provisions and Title V permitting requirements become applicable to what was previously defined as an area source category.

Section 112(c)(6) List of Specific Pollutants

Section 112(c)(6) requires that by 1995, sources accounting for not less than 90 percent of the aggregate emissions of each of seven specific pollutants must be listed on the source category list, and be subject to standards under 112(d)(2) or (4) no later than 2005. The pollutants are: alkylated lead compounds; polycyclic organic matter; hexachlorobenzene; mercury; polychlorinated biphenyls; 2,3,7,8-tetrachlorodibenzo-p-dioxin; and 2,3,7,8-tetrachlorodibenzofuran. This provision also includes a specific reference to utility boilers. It reads: "This paragraph shall not be construed to require the Administrator to promulgate standards for such pollutants emitted by electric steam generating units."

Section 112(d) Emission Standards

Section 112(d) requires that emission standards be established for each source category listed on the source category list. The emission standards are applicable to both new and existing sources and are based on the application of MACT. MACT is defined differently for new and existing sources as explained by 112(d)(2) and (3). Under 112(d)(4), if the pollutant is a threshold pollutant (e.g., noncarcinogen), the emission standard can be based on a health threshold with an ample margin of safety. A health threshold is a level where the risk of an adverse effect from exposure to the pollutant is negligible. Section 112(d)(5) allows the Administrator the discretion to apply generally available control technology (GACT) to area sources rather than MACT (or any other technologies that may be required of the source category on account of residual risk analyses under 112(f)).

Section 112(f) Residual Risk Program

Section 112(f) requires the U.S. EPA to report to Congress on the methods that will be used to calculate the risk remaining after the promulgation of MACT emission standards under Section 112(d). This report should address the public health significance of the risk and the actual health effects experienced by persons living in the vicinity of emitting sources, and make recommendations on legislation regarding the risk. This report is due to Congress on November 15, 1996. If Congress does not accept any of the recommendations provided for reducing the residual risk, the Administrator has the authority to promulgate any additional standards required in order to protect public health with an ample margin of safety. The report is currently under development.

Section 112(k) Urban Area Source Program

By 1995, a national strategy to control emissions of hazardous air pollutants (HAPs) from area sources in urban areas is required to be transmitted to Congress. The strategy must identify not less than 30 HAPs which present the greatest threat to public health in the largest number of urban areas. Source categories accounting for at least 90 percent of the aggregate emissions of each HAP must be listed on the source category list and be subject to 112(d) standards. The strategy, when implemented, is to achieve a 75 percent reduction in cancer incidence attributable to these sources.

Mercury is a likely candidate for the urban area source program.

112(m) Atmospheric Deposition to Great Lakes and Coastal Waters (Great Waters)

The Great Waters study is an ongoing study with periodic reports to Congress required. This program must identify and assess the extent of atmospheric deposition of HAPs to the Great Waters, the environmental and public health effects attributable to atmospheric deposition and the contributing sources. The first report was submitted in May 1994 and is to be submitted biennially hereafter. Mercury was identified as a priority pollutant under the Great Waters program. The Administrator must determine if other provisions under Section 112 will adequately control these sources. If not, by 1995, further emission standards to control these sources must be promulgated.

The recommendations of the first Great Waters Report to Congress included (1) the U.S. EPA should strive to reduce emissions of the identified pollutants of concern, including mercury, through implementation of the CAA; (2) a comprehensive approach should be taken both within the U.S. EPA and between the U.S. EPA and other Federal agencies to reduce and preferably prevent pollution in the air, water and soil; and (3) the U.S. EPA should continue to support research for emissions inventories, risk assessment and regulatory benefits assessment.

112(n)(1)(A) Study of Hazardous Air Pollutants for Electric Utility Steam Generating Units

The Utility Study is required to address the hazards to public health that are reasonably anticipated to occur as a result of emissions by electric utility steam generating units of ... [hazardous air pollutants] ... after imposition of the requirements of the Act. The list of 189 HAPs is presented in section 112(b) of the CAA. In the study, the U.S. EPA must develop and describe alternative control strategies for HAPs that may require regulation under section 112, and, if appropriate and necessary, the U.S. EPA is to proceed with rulemaking to control HAP emissions from utility boilers. Mercury is one of the pollutants of concern for utilities.

Section 129 Solid Waste Combustion

Under this section, the Administrator must establish emission guidelines and standards for solid waste incineration units, including municipal waste combustors, medical waste incinerators and commercial and industrial waste incinerators. The performance standards must specify numerical emission limits for mercury as well as a number of other pollutants. The U.S. EPA has already issued final rules for municipal waste combustors (59 FR 48198) and proposed rules for medical waste incinerators (60 FR 10654). Emission limits for hazardous waste combustors will be forthcoming under the Agency's Combustion Strategy.

Ongoing Activities

The U.S. EPA already has efforts underway to reduce mercury emissions from industrial sources. Specific actions being taken under the Clean Air Act include the following:

- The U.S. EPA has promulgated final emission limits for municipal waste combustors and is pursuing promulgation of proposed emission limits for medical waste incinerators under the authority of section 129 of the CAA.
- The U.S. EPA is evaluating the impacts of mercury reductions for the following source categories under the authority of section 112(c)(6): commercial/ industrial boilers, primary lead smelters, primary copper smelters, chlor-alkali plants using the mercury cell process and portland cement kilns.
- The U.S. EPA plans to evaluate whether secondary mercury production should be added to the source category list under section 112(c) of the CAA and subsequently evaluated for regulation under the authority of section 112(c)(6).
- Numerous CAA requirements involve utilities either directly or indirectly. Section 112(n)(1)(B) which required this Mercury Study Report to Congress specified utility boilers for analysis as did section 112(n)(1)(A) which is referred to as the Utility Air Toxics Report to Congress (Utility Study). The Utility Study is charged with evaluating the hazards to public health reasonably anticipated to occur as a result of emissions by electric utility steam generating units of pollutants listed under Section 112(b), including mercury, and to evaluate the impact of other provisions of the CAA on these emissions. The other provisions of the CAA would include the Acid Rain program as well as provisions pertaining to National Ambient Air Quality Standards. The Utility Study is also required to offer a regulatory recommendation with respect to regulation of utility boilers under section 112 of the CAA.

To address cross-media issues, additional pollution prevention options and regulatory authorities, the U.S. EPA has established a Mercury Task Force to consider strategies for coordinating various programs for use, management and disposal of mercury. The Task Force has recommended to the Department of Defense that the Defense Logistics Agency suspend sales of mercury from federal stockpiles through the fiscal year 1996 sales cycle, pending development of an EPA strategy for mercury. The U.S. EPA will make a final recommendation on the stockpile sales as part of this overall strategy.

During 1995 and beyond, the Mercury Task Force will consider several approaches for reducing mercury releases and environmental and human health risks associated with mercury exposure. The U.S. EPA will examine a wide range of options, within a multi-media framework, advocating common-sense pollution prevention programs. Some areas which the Task Force will explore include evaluation and

information transfer of ongoing prevention and control efforts at local, national and international levels; consideration of pollution prevention ideas including product substitution and innovation; recycling and disposal options; research and science needs; and coordination within U.S. EPA for consistent mercury regulatory programs, as well as coordination with Federal agencies managing mercury.

The findings of the Mercury Study Report to Congress will be considered by the Mercury Task Force as it develops a strategy.

5. RESEARCH NEEDS

The following sections summarize the major research needs identified for each of the study areas addressed in this Report.

Anthropogenic Mercury Emissions in the United States

An effort has been made to characterize the uncertainties (at least qualitatively) in the emissions estimates for the various source categories described. There are inherent uncertainties in estimating emissions using emission factors. To reduce these uncertainties, a number of research needs remain, including the following.

1. Source test data are needed from a number of source categories that have been identified as having insufficient data to estimate emissions. Notable among these are mobile sources, landfills, agricultural burning, sludge application, coke ovens, petroleum refining, residential woodstoves, mercury compounds production and zinc mining. A number of manufacturing sources were also identified as having highly uncertain emissions estimates. Notable among this category are secondary mercury production, commercial and industrial boilers, electric lamp breakage, primary metal smelting operations and iron and steel manufacturing. The possibility of using emissions data from other countries could be further investigated.
2. Development and validation of a stack test protocol for speciated mercury emissions is needed.
3. More data are needed on the efficacy of coal cleaning and the potential for slurries from the cleaning process to be a mercury emission source.
4. More data are needed on the mercury content of various coals and petroleum and the trends in the mercury content of coal burned at utilities and petroleum refined in the U.S.
5. Additional research is needed to address the potential for methylmercury to be emitted (or formed) in the flue gas of combustion sources.
6. The importance (quantitatively) of re-emission of mercury from previously deposited anthropogenic emissions and mercury-bearing mining waste needs to be investigated. This would include both terrestrial and water environments. Measuring the flux of mercury from various environments would allow a determination to be made of the relative importance of re-emitted mercury to the overall emissions of current anthropogenic sources.
7. Determination of the mercury flux from natural sources would help determine the impact of U.S. anthropogenic sources on the global mercury cycle as well as the impact of all mercury emissions in the United States.
8. The use of more sophisticated fate and transport models for mercury will require more detailed emissions data, particularly more information on the chemical species of mercury being emitted (including whether these species are particle-bound) and the temporal variability of the emissions.

Exposures from Anthropogenic Mercury Emissions in the United States

During the development of the mercury exposure assessment, many areas of uncertainty and significant data gaps were identified. Many of these have been identified in the document, and several are presented in the following list.

1. Improved analytical techniques for measuring speciated mercury air emissions are needed as well as total mercury emissions from point sources. Laboratory evidence suggests that divalent mercury gas emissions will wet and dry deposit much more readily than elemental mercury gas. Particle-bound mercury is also likely to deposit relatively quickly. Current stack sampling methods do not provide sound information about the fraction of mercury emissions that are in oxidized form. While filters are used to determine particulate mercury fractions, high temperature stack samples may not be indicative of the fraction of mercury that is bound to particles after dilution and cooling in the first few seconds after emission to the atmosphere. Methods for determination of the chemical and physical forms of mercury air emissions after dilution and cooling need to be developed and used to characterize significant point sources.
2. Evaluated local and regional atmospheric fate and transport models are needed. These models should treat all important chemical and physical transformations which take place in the atmosphere. The development of these models will require comprehensive field investigations to determine the important atmospheric transformation pathways (e.g., aqueous cloud chemistry, gas-phase chemistry, particle attachment, photolytic reduction) for various climatic regions.
3. The evaluation of these models will require long-term national (possibly international) monitoring networks to quantify the actual air concentrations and surface deposition rates for the various chemical and physical forms of mercury.
4. Better understanding of mercury transport from watershed to water body including the soil chemistry of mercury, the temporal aspects of the soil equilibrium and the impact of low levels of volatile mercury species in surface soils and water bodies on total mercury concentrations and equilibrium.
5. Better understanding of foliar uptake of mercury and plant/mercury chemistry. (The most important questions: do plants convert elemental or divalent mercury into forms of mercury that are more readily bioaccumulated? Do plants then emit these different forms to the air?) A better understanding of the condensation point for mercury is needed.
6. Better understanding of mercury movement from plant into soil (detritus). May need to refine the models used to account for movement of mercury in leaf litter to soil.
7. The impact of anthropogenic mercury on the "natural," existing mercury levels and species formed in soil, water, and sediments needs better understanding. How does the addition of anthropogenic mercury affect "natural" soil and water mercury cycles? Natural emission sources need to be studied better and their impacts better evaluated.
8. Improved understanding of mercury flux in water bodies and impact of plant and animal biomass are needed. Unlike many other pollutants, most of the methylmercury in a water body appears to be in the biological compartment. The sedimentation rate as well as benthic sediment:water partition coefficient require field evaluation. Important to consider rivers and other larger water bodies in these flux analyses.

9. The bioaccumulation factors (BAF) contains a substantial level of uncertainty. A more appropriate BAF can probably be developed when the data base upon which the estimate is based is enlarged; i.e., need data from more than four studies. The availability of more data would enable the possible development of lake-type adjustment factors for the mercury BAF possibly based on color, acidification susceptibility, etc., or species-specific BAF adjustment factors for freshwater species most commonly consumed. Also need a time analysis of fish mercury uptake which could lead to the development of a dynamic fish model. A mercury BAF for marine fish is needed.
10. Better estimates of fish consumption rates for high-end consumers (subsistence) as well as recreational anglers are needed. Fish species-specific consumption rates are also needed. Improving these estimates would require additional dietary surveys.
11. Need to improve the biotransfer factors for mercury from soil and plants to beef.
12. Long-term studies using ultra-clean sampling techniques and state-of-the-art analytical methods are needed to help resolve questions of mercury concentration trends over time, particularly in soils, sediments and biota.
13. A research need for this area is for biological monitoring (for exposure and effect) of populations with either greater than U.S. average fish consumption (such as one serving of 100 grams per day) or consumption of fish predicted or measured to have higher than average amounts of methylmercury.

Health Effects of Mercury and Mercury Compounds

1. In addition to the ongoing studies identified in the health effects review, further research is necessary for refinement of the U.S. EPA's risk assessments for mercury and mercury compounds. In order to reduce uncertainties in the current estimates of the oral reference doses (RfDs) and inhalation reference concentrations (RfCs), longer-term studies with low-dose exposures are necessary. In particular, epidemiological studies should emphasize comprehensive exposure data with respect to both dose and duration of exposure. Some studies should be targeted to populations identified in this Report as likely to experience methylmercury exposure in fish (e.g., subsistence fishers).
2. The current RfD and RfC values have been determined for the most sensitive toxicity endpoint for each compound; that is, the neurological effects observed following exposure to elemental or methylmercury, and the renal autoimmune glomerulonephritis following exposure to inorganic mercury. For each of these compounds, experiments conducted at increasingly lower doses with more sensitive measures of effect will improve understanding of the respective dose-response relationships at lower exposure levels and the anticipated thresholds for the respective effects in humans. Similar information from developmental toxicity studies would allow determination of RfDs for developmental toxicity (RfD_{dt}) for elemental and inorganic mercury.
3. Research needs include studies which will delineate the most appropriate indicators of neurotoxic effects for exposed adults, children and individuals exposed to methylmercury *in utero*. Well conducted studies are also needed to clarify critical levels at which other toxic effects could occur in humans.

4. Well-conducted studies are also needed to clarify exposure levels at which toxic effects other than those defined as “critical” could occur in humans. For all three forms of mercury, data are inadequate, conflicting, or absent for the following: adverse reproductive effects (effects on function or outcome, including multigeneration exposure); impairment of immune function; and genotoxic effects on human somatic or germinal cells (elemental and inorganic mercury).
5. Investigations that relate the toxic effects to biomonitoring data will be invaluable in quantifying the risks posed by these mercury compounds. In addition, work should focus on subpopulations that have elevated risk because they are exposed to higher levels of mercury at home or in the workplace, because they are also simultaneously exposed to other hazardous chemicals, or because they have an increased sensitivity to mercury toxicity.
6. There are data gaps in the carcinogenicity assessments for each of the mercury compounds. The U.S. EPA's weight-of-evidence classification of elemental mercury (Group D) is based on studies in workers who were also potentially exposed to other hazardous compounds including radioactive isotopes, asbestos, or arsenic. There were no appropriate animal studies available for this compound. Studies providing information on the mode of action of inorganic mercury and methylmercury in producing tumors will be of particular use in defining the nature of the dose response relationship.
7. The assessment of both noncarcinogenic effects and carcinogenic effects will be improved by an increased understanding of the toxicokinetics of these mercury compounds. In particular, quantitative studies that compare the three forms of mercury across species and/or across routes of exposure are vital for the extrapolation of animal data when assessing human risk. For elemental mercury there is a need for quantitative assessment of the relationship between inhaled concentration and delivery to the brain or fetus; in particular the rate of elemental to mercuric conversion mediated by catalase and the effect of blood flow. Such assessment is needed for evaluation of the impact of mercury exposure from dental amalgam.
8. Work has been done on development of physiologically-based pharmacokinetic models. While one of these has developed a fetal submodel, data on fetal pharmacokinetics are generally lacking. The toxicokinetics of mercury as a function of various developmental stages should be explored. Elemental mercury and methylmercury appear to have the same site of action in adults; research is, therefore, needed on the potential for neurotoxicity in newborns when the mother is exposed. This work should be accompanied by pharmacokinetic studies and model development.

Ecological Assessment for Anthropogenic Mercury Emissions in the United States

1. *Process-based Research.* Mechanistic information is needed to understand the variability that presently typifies the mercury literature. This research includes laboratory and field studies to identify the determinants of mercury accumulation in aquatic food chains and kinetic information that would allow researchers to describe the dynamics of these systems. Areas of uncertainty include these: (1) factors that determine net rates of methylation and demethylation; (2) dietary absorption efficiency from natural food sources; (3) effect of dietary choice; and (4) bioavailability of methylmercury in the presence of dissolved organic material and other potential ligands.

In time it is anticipated that this information can be used to develop process-based models for mercury bioaccumulation in fish and other aquatic biota. Significant progress in this direction is represented by the Mercury Cycling Model (MCM), presently being developed and evaluated by the Electric Power Research Institute (Hudson et al., 1994).

2. *Wildlife Toxicity Data.* There is a need to reduce the present reliance on a relatively few toxicity studies for WC development. Additional data are needed for wildlife that constitute the most exposed organisms in various parts of the country (e.g., the Florida panther). There is also a critical requirement for toxicity data that can be related to effects on populations (see Table 2-1), including effects on organisms that comprise the lower trophic levels.
3. *Improved Analytical Methods.* Efforts to develop and standardize methods for analysis of total mercury and methylmercury in environmental samples should be continued. Such methods must recognize the importance of contamination, both during the collection of such samples and during their analysis. It is particularly important that mercury measurements which are at present operationally defined (e.g., "soluble", "adsorbed to organic material") be made in such a way that mercury residues in fish can be correlated with the bioavailable mercury pool.

As validated methods become available, it is important to analyze for both total and methylmercury whenever possible so that differences between aquatic systems can be definitively linked to differences in methylmercury levels. Analyzing the two mercury species together will contribute to an understanding of existing data, most of which is reported as total mercury. It is also anticipated that developing BAFs in terms of methylmercury will reduce the variability that currently exists around BAF estimates based on total mercury.

4. *Complexity of Aquatic Food Webs.* Present efforts to develop WC values for mercury are based on linear, four-tiered food chain models. Research is needed to determine the appropriateness of this simple paradigm and to develop alternatives if field data suggest otherwise. Of particular interest is whether zooplankton and phytoplankton should be modeled as two different trophic levels. Current information for detritivores and benthic invertebrates is extremely limited, even though their importance in mobilizing hydrophobic organic contaminants has been demonstrated.
5. *Accumulation in Trophic Levels 1 and 2.* Ongoing efforts to understand mercury bioaccumulation in aquatic systems continue to be focused on trophic levels 3 and 4, despite the fact that uncertainties in predator/prey factors (PPFs) are relatively small. Additional emphasis should be placed on research at the lower trophic levels. In

particular, there is a need to understand the determinants of mercury accumulation in phytoplankton and zooplankton, and how rapid changes in plankton biomass impact these values.

6. *Field Residue Data.* High quality field data are needed to support process-based research efforts and to determine residue concentrations in the fish and other aquatic biota that wildlife eat. Whenever possible, it is desirable to collect residue data at all trophic levels and to analyze mercury levels in the abiotic compartments of a system (e.g., water and sediments). It is particularly important that such measurements be made in a broader array of aquatic ecosystem types (including both lakes and rivers) so that a better understanding of mercury cycling and accumulation can be obtained.

Residue data from wildlife are also needed to identify populations that are being adversely impacted or are potentially at risk. Feathers and fur hold considerable promise in this regard because of the potential for "non-invasive" determination of mercury residues. Laboratory research is required, however, to allow interpretation of these data. Factors such as age, sex, and time to last moult are likely to result in variability among individuals of a single population, and need to be understood. Sampling efforts should be targeted on areas receiving high levels of mercury deposition and/or regions containing large numbers of poorly buffered surface waters, as discussed throughout this report.

7. *Natural History Data.* The development of WC requires knowledge of what wildlife eat. Fish sampling efforts are frequently focused on species that are relevant to human consumers but that may be of little significance to wildlife. There is an additional need to collect information for macroinvertebrates and amphibians. Seasonal and spatial effects on predation should be explored and methods developed to describe this information adequately. Additional life history data is needed to characterize fully the nature and extent of exposure to mercury. Complicating factors must be considered, including migratory behaviors and sex-specific differences in distribution and resource allocation. It is particularly important that information be collected to support the development of predictive population models for sensitive species.

Risk Characterization

1. A monitoring program is needed to assess either blood mercury or feather/hair mercury of piscivorous wildlife; particularly those in highly impacted areas. This program should include assessment of health endpoints including neurotoxicity and reproductive effects.
2. There is a need to collect additional monitoring data on hair or blood mercury and assess health endpoints among women of child-bearing age and children. This study should focus on high-end fish consumers and on consumption of fish from contaminated water bodies.
3. There is a need for improved data on effects that influence survival of the wildlife species as well as on individual members of the species.
4. There is a need for controlled studies on mercury effects in intact ecosystems.
5. Monitoring data sufficient to validate or improve the local impact exposure models are needed.

Mercury Control Technologies

1. Data are needed from full-scale testing of activated carbon injection at a coal-fired utility boiler.
2. Additional data are needed on the efficiency of activated carbon injection, and various impregnated carbons, in reducing the different chemical species of mercury present in flue gas.
3. Additional information is needed on the efficiency and cost of other technologies for mercury control that are currently in the research stage. These include impregnated activated carbon, sodium sulfide injection and activated carbon fluidized bed.
4. More data are needed on the ability of conventional or advanced coal cleaning techniques to remove mercury from raw coal and advanced coal-cleaning techniques such as selective agglomeration and advanced column floatation. The potential for mercury emissions from coal-cleaning slurries need to be characterized.
4. Additional analyses are required on the feasibility and cost effectiveness of other mercury emission prevention measures such as emissions trading, emissions averaging, energy conservation, renewable energy, and fuel switching.

APPENDIX A
SUMMARY OF THE SCIENTIFIC PEER REVIEW

1. OVERVIEW OF THE REVIEW PROCESS

Ensuring the quality of scientific underpinning actions by U.S. EPA has been a major thrust of Agency policy. In order to ensure top quality science, U.S. EPA has been advised by review bodies including its Science Advisory Board (SAB) to consider peer review of its programs, methods and products to be a top priority. To this end Administrator Carol Browner issued a memorandum requiring expert review wherever appropriate and requiring U.S. EPA Programs and the Office of Research and Development (ORD) to develop specific guidelines for scientific review. The ORD final guidelines for review of scientific products were issued in November 1994. These were used as the basis for the peer review plan for this Report. The Mercury Study Report to Congress was considered by ORD and the Office of Air Quality Planning and Standards, Office of Air and Radiation (OAQPS/OAR) to be one of U.S. EPA's major and most visible outputs. As such the Report was considered to fall into category 1, requiring the highest level of scientific peer review. The components for category 1 review include the following: approval of the peer review plan by the Assistant Administrator of ORD; review of the product by appropriate U.S. EPA scientists; review of the product by appropriate scientists external to the Agency; convening a peer review meeting; and stringent recordkeeping on all phases of the review process.

Because of the wide scope of the Report and the interest in mercury by many stakeholders, it was felt that the process of generating the Report should be open to external input. Meetings with U.S. EPA Report authors were held with members of the public at their request; for example, during early stages of Report generation, U.S. EPA staff met on a quarterly basis with scientists and engineers representing the Electric Power Research Institute (EPRI). Meetings were also held with the Portland Cement Association and with other requestors. The Agency accepted and reviewed submissions of data and mercury assessment material throughout the study period; these were used as was considered appropriate by U.S. EPA scientists.

In order to gather input and critiques on preliminary assessments, several of these were presented at conferences and scientific meetings. Early results of the emissions inventory (found in Volume II) were presented at both regional and national meetings. Draft health assessments were also shown for purposes of discussion at scientific meetings on mercury. In January of 1994 a review draft of the emissions inventory was made publicly available.

Internal scientific review of a draft of the entire Report (minus Volume I, Executive Summary) was begun in November of 1994. Following procedures for review and clearance established for the Office of Health and Environmental Assessment (now the National Center for Environmental Assessment, NCEA) within ORD, the draft was reviewed by four scientists from that Office. In addition, the draft was reviewed by a U.S. EPA Mercury Study Work Group consisting of staff from the following U.S. EPA offices: Office of Science, Planning and Regulatory Evaluation (OSPRE/ORD); Office of Health Research (OHR/ORD); Office of Policy Analysis and Review (OPAR/OAR); Office of Water (OW); Office of Solid Waste and Emergency Response (OSWER); Office of Prevention, Pesticides and Toxic Substances (OPPTS); Office of Policy, Planning and Evaluation (OPPE); and Region V. Scientists representing the State of New York and the State of Michigan also participated in the Work Group and in this phase of review.

Included as part of the Report are summaries of human health risk assessments which comprise parts of the Agency's Integrated Risk Information System (IRIS). IRIS is a publicly available computerized data base which provides U.S. EPA consensus health risk assessment information. IRIS files must undergo specific forms of internal and external review before they are made available on the system. The IRIS documents on mercury were reviewed as part of the Mercury Study Report to Congress. The following six IRIS summaries were distributed to reviewers as Appendix B of Volume IV of the Report: reference concentration for elemental mercury; cancer assessment for elemental mercury;

reference dose for inorganic mercury; cancer assessment for inorganic mercury; reference dose for methylmercury; and cancer assessment for methylmercury. Internal review for the IRIS documents consisted of the appropriate Agency Work Group discussion and closure (referred to as "verification"). The Work Groups charged with reviewing IRIS information and achieving consensus on its validity are comprised of U.S. EPA scientists from a variety of disciplines relevant to human health risk assessment and who represent ORD, the Regions and the Program Offices. The two Work Groups have been organized around either the assessment of carcinogenic effects (the Carcinogen Risk Assessment Verification Endeavor (or CRAVE) or the assessment of general systemic toxicity (RfD/RfC) Work Group). To enhance the mercury expertise of the Work Groups and to allow for discussion of alternate risk assessment approaches, scientists from FDA, ATSDR and the State of New Jersey were invited to participate in the RfC/RfD Work Group discussions; they were not part of the consensus process, however, and did not participate in Agency decisions on the assessments. After consensus on the assessment was achieved, IRIS documents were revised and received external review (see below) as part of the external review draft of the Report. Following external review and revision the IRIS documents were either reviewed and cleared by the Work Group chair (RfDs and RfC) or given a pass-around review by the whole Work Group (CRAVE).

External review of the Mercury Report to Congress and the appended IRIS documents was done in two steps: a Federal interagency review and a non-Federal external review. A meeting of Federal reviewers was held at the U.S. EPA, Washington DC on January 9, 1995 to discuss scientific issues in the Report. Representatives of the following Agencies were invited to attend and to submit written comments at the time of the meeting: ATSDR, NIEHS, NOAA, USDA, DOE, FDA and the National Biological Service. The names and addresses of the reviewers can be found at the beginning of each volume of the Report. Written comments were received from all Agencies participating in the review. A summary of reviewer comments, consensus opinion of reviewers and U.S. EPA's response to comments can be found in this Appendix.

The second phase of external review included comments from non-Federal experts. Reviewers were chosen based on scientific expertise and availability. An attempt was made to include representatives of a spectrum of groups with interest in mercury: academe, research groups, State agencies, industrial concerns and environmental groups. The names and affiliations of reviewers can be found at the beginning of each volume. All reviewers were required to submit written comments on the report including the IRIS documents. A public review meeting was held January 25-26, 1995 at U.S. EPA in Cincinnati, OH. A notice of the meeting was published in the Federal Register, and there was time set aside each meeting day for members of the public to comment. The pre- and post-meeting reviewer comments, synopses of meeting discussions and conclusions of the meeting comprised the external review report. Examples of external review comments are in Appendix A to this Volume. In response to reviewer comments, specific changes were made in the External Review Draft. As the Risk Characterization (Volume VI) was revised, a second review of that volume was done.

One revised component was reviewed in advance of the remainder of Volume VI; this was the estimate of population size, amount of fish consumed and measured amount of mercury in marine and freshwater fish. This assessment (now included in Volume III, Appendix H and summarized in Volume VI) was sent to two external reviewers expert in statistics and demographics. These reviewers were selected by a U.S. EPA contractor who was provided with criteria for reviewers and a list of potential candidates. The entire revised Risk Characterization was subjected to internal and external review. Scientists in ORD, OAQPs and Office of Water were sent the volume and requested to submit comments. Four external reviewers were selected by a U.S. EPA contractor based on criteria provided by U.S. EPA. Among these criteria were that two reviewers be included who had commented on the External Review draft. Written comments on the risk characterization were provided by these four external scientific

reviewers. Copies of all review comments and external input are archived at the National Center for Environmental Assessment in Cincinnati, Ohio.

This appendix summarizes the major comments provided by external and Federal interagency reviewers, along with U.S. EPA's responses. Section 2 presents an overview of the charge to the external and interagency reviewers. Section 3 provides a summary of the external review process, the non-Federal, external reviewers comments and U.S. EPA's disposition. Finally, Section 4 summarizes U.S. EPA's notes and responses to comments from Federal interagency reviewers.

2. CHARGE TO REVIEWERS

Reviewers were asked to focus on that portion of the report that matched their area of expertise. The following are issues or questions considered by the reviewers, including the development of premeeting comments.

All Volumes

- Are additional data or analyses available that would have a major impact on the conclusions presented in any volume of the report?
- Are arguments and conclusions presented clearly and in a logical manner?
- Do the Research Needs chapters of particular volumes present a program of research projects that will address uncertainties in the evaluation of mercury impacts?

Volume I: Executive Summary

- Does the summary adequately reflect the conclusions of the other volumes?
- Is additional information presented in the report that should be added to the summary for clarity or completeness?
- Is the summary sufficiently clear and informative to function as a stand-alone volume, or does it rely too heavily on familiarity with the report as a whole?

Volume II: An Inventory of Anthropogenic Mercury Emissions in the United States

- Please critique the emission factors approach used in the inventory.
- Are you aware of information for source categories identified as having insufficient data for evaluation?

Volume III: An Assessment of Exposure From Anthropogenic Mercury Emissions in the United States

- Please critique the conclusions of the exposure modeling. Are the conclusions well supported by the analyses presented in the text of Volume III?
- Is there material in the text of Volume III that would be more appropriately presented in an appendix?
- Please critique methods used and assumptions made for the local impact analysis.
- Do the appendices provide necessary supporting information concerning methods described in the text?

Volume IV: Health Effects of Mercury and Mercury Compounds

- Is the information provided on pharmacokinetics sufficient for evaluating human health effects associated with mercury?
- Please critique the weight-of-evidence categorizations for carcinogenicity, developmental toxicity, and germ cell mutagenicity. Is the level of detail in the report descriptions in Volume IV sufficient to permit evaluation of these endpoints?
- No quantitative dose-response assessment was conducted on carcinogenicity for inorganic or methyl mercury. Are the arguments against conducting a quantitative assessment presented cogently and are they supported by the information given in this volume?
- Are the reference doses (RfDs) and reference concentrations (RfCs) properly calculated? Were the appropriate critical study and endpoint(s) chosen? Were the proper uncertainty factors and modifying factors used?
- Are there any factors modifying mercury toxicity in humans that have not been addressed in the volume?

Volume V: An Ecological Assessment for Anthropogenic Mercury Emissions in the United States

- Please critique the methods used for generating a trophic level three BAF and a trophic level four BAF.
- Please critique the methods used for generating an uncertainty analysis.
- Were appropriate endpoints and studies selected for generating wildlife RfDs?
- Were appropriate assumptions used in developing wildlife water criteria?
- Are there other species of concern that should be considered in this volume?
- Are there other geographic areas of concern that should be included in this volume?

Volume VI: Characterization of Human Health and Wildlife Risks From Anthropogenic Mercury Emissions in the United States

- Are the summaries of human and wildlife risk assessment sufficient for a scientific critique?
- Are there major areas of uncertainty, defaults, or assumptions that were not discussed?
- Please critique the uncertainty analyses.
- Please critique both the methods and results of the comparative discussion of risk presented in this volume.

Volume VII: An Evaluation of Mercury Control Technologies and Costs

- Are you aware of any quantified benefits of mercury control? Please specify.
- Are you aware of data on the efficacy of materials separation programs or other pollution prevention measures other than that presented in this volume? Please specify.
- Please critique the cost analysis presented in this volume.

3. SUMMARY OF NON-FEDERAL EXTERNAL REVIEWERS COMMENTS AND U.S. EPA DISPOSITION

On January 25-26, 1995, a 1½-day workshop was held at the U.S. EPA's Andrew W. Breidenbach Environmental Research Center in Cincinnati, Ohio, to provide external review of the draft *Mercury Report to Congress*. A draft report was prepared by U.S. EPA's Office of Air Quality Planning and Standards and Office of Research and Development in response to Section 112(n)(1)(B) of the Clean Air Act Amendments of 1990, which requires U.S. EPA to submit a report to Congress on mercury emissions. The draft report consisted of six volumes at the time it was distributed for review:

- Volume II: An Inventory of Anthropogenic Mercury Emissions in the United States.
- Volume III: An Assessment of Exposure from Anthropogenic Mercury Emissions in the United States.
- Volume IV: Health Effects of Mercury and Mercury Compounds.
- Volume V: An Ecological Assessment for Anthropogenic Mercury Emissions in the United States.
- Volume VI: Characterization of Human Health and Wildlife Risks from Anthropogenic Mercury Emissions in the United States.
- Volume VII: An Evaluation of Mercury Control Technologies and Costs.

Volume I, the Executive Summary, was not yet complete and ready for review along with the other volumes. In preparation for the workshop, Eastern Research Group, Inc., providing contractor support to U.S. EPA, identified 15 independent external scientists to review the document. The reviewers' expertise covered a variety of subject areas relevant to the report, including mercury emissions and sources of mercury emissions; the transport to and fate of mercury in the environment; the physicochemical and biotic transformation among mercury forms in environmental compartments, particularly of inorganic to methylmercury; exposure of human and ecological populations to methylmercury and other mercurials; human and ecological toxicology; quantitative risk assessment; and risk management. Each reviewer was asked to focus on that portion of the report that matched his or her area of expertise. Reviewers prepared and submitted premeeting comments on the report prior to the workshop.

Fourteen reviewers,¹ 10 U.S. EPA representatives involved in writing and/or revising the mercury report, and 39 observers attended the workshop. The agenda included plenary sessions and breakout groups. The first day of the workshop began with a presentation, by the two breakout group chairs, of summaries of the reviewers' premeeting comments for Volumes II, III, IV, and V. The participants then broke into two groups — one to discuss Volumes II and III, and the other to discuss Volumes IV and V. During a plenary session at the end of the first day, the breakout group chairs presented a summary of their groups' discussions and observers commented on the report.

The second day of the workshop consisted of a half-day plenary session. Two reviewers presented a summary of the premeeting comments on Volumes VI and VII, and all the reviewers then discussed these two volumes. Following this discussion, additional observers presented their comments on the mercury report.

¹One of the 15 original reviewers was unable to attend.

A summary of the major comments provided by external reviewers along with U.S. EPA's responses is presented in the following sections: a report of the workshop chair (Section 3.1); summary of premeeting comments (Section 3.2); summary of breakout group discussions (Section 3.3); overview of reviewer discussion in the plenary session (Section 3.4); and the summary of revisions made in response to reviewers comments (Section 3.5).

3.1 Report of the Workshop Chair - Paul Mushak, Ph.D.

Overview

The draft report and reviewers' comments clearly show that, while we do not know nearly as much as we would like to about environmental mercury, we know a lot. In fact, we know more about environmental mercury than about most contaminant metals or metalloids of concern.

The principal challenge for both the authors and external reviewers of the draft report was to critically evaluate the problems associated with integrating what we do and do not know into a scientifically credible synopsis. One of these problems appears to be that the extensive database for mercury is mainly available as discrete blocks of information within various scientific disciplines, while the congressional mandate requires U.S. EPA to establish and quantify linkages between these blocks of data. For example:

- Information in one block tells us that the forms of mercury addressed in the draft report — particularly methylmercury — are intrinsically toxic, with a relatively high degree of toxicological potency to humans and various other biological receptors. The types of toxic responses known or anticipated in both ecological and human populations are qualitatively recognized.
- Information in a second block tells us that mercury is emitted to the environment from a variety of sources, and that one can generally determine the relative contribution of different anthropogenic mercury source categories.
- Information in a third block tells us that some fraction of the mercury emitted to the atmosphere from a point source will eventually be deposited by precipitation processes onto land and water bodies. Direct or indirect post-depositional processes not only will impart mobility to the contaminant but also will transform mercurial species.
- Information in a fourth block tells us that inorganic ionic mercury entering certain environmental compartments will undergo biomethylation to methylmercury, and that methylmercury will accumulate and biomagnify in the human food web, particularly in high-trophic-level predator fish. Data in this block also show that mercurial forms can contaminate several environmental media, depending on the exposure particulars.

These examples of what we know clearly indicate that the difficulties in synthesizing all this information into a coherent statement about the potential health and ecological risks posed by mercury in the United States are rooted in uncertainties about how to quantitatively link these blocks together. Areas of uncertainty include, for example:

- How much of current anthropogenic atmospheric emissions is deposited in various environmental compartments?

- What is the link between natural and anthropogenic mercury in terms of proportional contamination and subsequent impact?
- How much of this post-deposition mercury is converted to highly toxic methylmercury?
- How much of any increased toxicity risk associated with consumption of methylmercury-contaminated fish can be traced back to anthropogenic atmospheric emissions of mercury?

The draft report was variably successful in dealing with the numerous complexities, uncertainties, and data gaps connected with quantifying linkages. The essence of the reviewers' comments concerned whether the report under- or overstated these uncertainties, particularly with reference to risk characterization.

General Review Panel Assessments of the Report

In their comments before and during the workshop, the peer reviewers recommended revisions to strengthen the report's scientific credibility. Reviewers generally agreed that the report would serve a useful purpose once it had been revised and improved in the various ways they had suggested. Few, if any, reviewers felt the report should not be submitted at all, and no reviewer thought the report should be transmitted without revision.

The review panel generally agreed that some portions of the report underestimated the uncertainty associated with modeled estimates or pathway analyses. The panel suggested that one way to better acknowledge this higher uncertainty was to use a range of values rather than point estimates in the estimating exercise; some panel members also argued that a more refined point estimate could be presented in certain cases—for example, in deriving the reference dose (RfD) for methylmercury.

On the other hand, the panel also generally agreed that the draft report conveyed too much uncertainty by failing to include important peer-reviewed data available in the recent scientific literature. For example, the exposure breakout group generally agreed that data do exist to indicate a relationship between point-source mercury emissions and gradients in mercury deposition consistent with a point-source contribution.

The review panel was similarly concerned about including or excluding available information on other topics in the report. The panelists felt that the authors should revisit the most recent scientific information to close any gaps that affect quantification of the linkages noted above.

Reviewers also were concerned about the role of modeling in the report. However, they had different opinions about how much data from the recent literature could be used to complement the model estimates. Panelists generally agreed that the report volumes should be more consistent and integrated, particularly concerning information relevant to risk characterization.

3.2 Summary of Premeeting Comments

This section presents the summary of the premeeting comments for the following Volumes of the draft Mercury Study Report to Congress:

- Volume II (Emissions) and Volume III (Exposure)—Gerald Keeler, Ph.D., and Paul Mushak, Ph.D;

- Volume IV (Health Effects)— Steven Bartell, Ph.D., and Paul Mushak, Ph.D;
- Volume V (Ecological Effects)— Steven Bartell, Ph.D., and Paul Mushak, Ph.D; and
- Volume VI (Risk Characterization)— Pamela Shubat, Ph.D., and Paul Mushak, Ph.D.

3.2.1 Volume II (Emissions) and Volume III (Exposure)— Gerald Keeler, Ph.D., and Paul Mushak, Ph.D.

Reviewers felt that Volume II probably was the best of the four volumes reviewed. The approach used to characterize emissions was reasonable. However, the volume provides no estimates of natural and baseline emissions and ignores several potentially important sources. Specific comments on the various sections are provided below.

Natural Emissions

The inadequate coverage of natural sources of mercury detracts from the entire report. Chapter 2, *Natural Sources of Mercury Emissions*, which consists of only a single page in Volume II, is incomplete and misleading. The topic of natural sources of mercury is controversial and qualitative at best. If the authors want to include this topic in the report, they should provide a more complete and defensible assessment of natural emissions. Reviewer William Fitzgerald, Ph.D., recommended that natural emissions could be roughly calculated using an approach similar to that of Mason et al. (1994).² This approach suggests that natural emissions in the United States are approximately 20 percent of anthropogenic emissions. A recent estimate of natural emissions in Europe gave a similar result of 25 percent of the total emissions (Axenfeld et al., 1992).³ However, the quantitative data concerning natural emissions are very limited, and there are numerous problems with the estimates in the literature.

Anthropogenic Sources

The report's list of source categories for mercury emissions is complete with respect to the major source categories. Many of the source categories discussed have relatively low annual mercury emissions. For a few source categories for which insufficient information was found, the report provides no emission estimates. Emission factors and data are missing for several potentially important sources, including hazardous waste incinerators, primary mercury production, mercury compounds production, by-product coke production, refineries, and mobile sources. In addition, Volume II provides no information or discussion on emissions from iron-steel production and primary zinc production. Emission factors and data are available for European sources and could be used to estimate the U.S. emissions to determine their potential importance.

The report to Congress provides only very limited information on emissions of various physical and chemical forms of mercury. Better information is needed on mercury speciation in both emissions and environmental samples. These could be identified as research needs.

The report could be strengthened by adding maps showing the actual location of point sources for categories like utilities (by fuel type), incinerators (sludge, municipal), iron-steel production, coke ovens,

²Mason, R.P., W.F. Fitzgerald, and F.M.M. Morel. 1994. Aquatic biogeochemical cycling of elemental mercury: anthropogenic influence. *Geochim. Cosmochim. Acta* 58:3191-3198.

³Axenfeld, F., J. Munch, and J.M. Pacyna. 1991. Europäische Test-Emissionsdatenbasis von Quecksilber-Komponenten für Modellrechnungen. Dornier Report. Friedrichshafen, Germany.

and cement production. The spatial distribution of the gridded emissions presented at the workshop by report author Martha Keating should also be included.

Lastly, the report lacks information regarding seasonal or temporal variations in emissions by source category. While utilities may have fairly constant emissions both diurnally and seasonally, other sources do not. Operations involving multiple steps over different time periods will probably have time-varying emissions.

Exposure to Mercury

A comprehensive quantitative assessment of the relationship between anthropogenic mercury releases to the atmosphere and the potential exposure of people, wildlife, and terrestrial and aqueous systems to these releases may not be possible due to the apparently limited state of knowledge of the mercury cycle in nature and the environmental consequences from anthropogenic emissions of mercury. The report states that the exposure assessment is a "qualitative study based partly on quantitative analyses." As noted by reviewer William Fitzgerald in his premeeting comments:

...this important exposure assessment provides a valuable guide for research. Although the results and conclusions are qualitative, this extensive and essential modeling effort provides a credible means for evaluating the present sparse data base, and for identifying major gaps, inconsistencies and weaknesses associated with major aspects of the biogeochemical cycle of Hg at the Earth's surface....

As the report confirms, human exposure to methylmercury is almost exclusively from consumption of fish and fish products. Intake of methylmercury through consumption of nonlocal fish and seafood should be evaluated. Such intake should not be considered "background," as the mercury found in coastal environments and in saltwater fish may be of anthropogenic origin. The report lacks an assessment of the exposure of the marine environment—especially the coastal zone—to anthropogenic mercury emissions and of the effects of such exposure.

The report suffers from a general lack of recent information and actual measurement data in the recent peer-reviewed literature. References will be provided by the reviewers, and the *Monterey Mercury Meeting Book* will be provided by Donald Porcella (Electric Power Research Institute). Inclusion of more recent information will address such comments as "There is a general recognition of uncertainty," "So much is said about uncertainty that it appears as if we do not know much about mercury," and "Little of the most recent knowledge has found its way into this report."

The modeling results should be "ground-truthed" where possible. The report's estimates of deposition and water concentrations often are more than an order of magnitude greater than any actually measured in the United States.

The meaning of some key terms used in the report, such as "total emissions" and "background," was confusing. The peer reviewers strongly recommended that the authors add to the report the definitions provided in the *Atmospheric Mercury Expert Panel Report* and that they use the various terms consistently throughout the report based on these definitions.

3.2.2 Volume IV (Health Effects)—Steven Bartell, Ph.D., and Paul Mushak, Ph.D.

Several of the reviewers' key premeeting comments concerned the major sources of intake and exposure in human populations. Some reviewers suggested that the report should address the contributions to human mercury exposure of dental amalgams containing mercury. Similarly, reviewers

recommended that the drinking water pathway be further examined, including the potential human health risks associated with drinking water at locations known to have elevated mercury concentrations in ground water. The report should clearly explain why particular papers concerning human health endpoints are cited while others were omitted.

Reviewers also commented on the subject of mercury disposition among biological indicators of mercury exposure, particularly exposure to methylmercury. The derivation and use of a constant ratio of mercury in hair to mercury in blood for estimating blood levels of mercury may require additional attention. Reviewers expressed reservations about the time-scale differences implicit in comparing blood mercury with hair mercury—namely, that mercury concentrations in hair reflect exposure over a longer time scale, while mercury concentrations in blood may correspond to a shorter time frame. The reported variability may reflect interindividual variability rather than just measurement error as Volume IV suggests. Reviewers identified an error in the equation used to calculate the methylmercury concentration in blood; an additional term defining blood volume is needed to make the units in this equation work out to those stated.

The quantitative linkage of mercury intake by exposed populations and the expression of some toxic endpoint is mediated through the toxicokinetics—i.e., the uptake, distribution, and retention/excretion—of the particular mercurial. The modeling of the systemic behavior of methylmercury is particularly critical in this regard. The reviewers felt that the derivation of the parameters used in the pharmacokinetic modeling needed additional explanation and justification. For example, the elimination rate or half-life used to describe methylmercury conversion to inorganic mercury and its subsequent removal from the body in feces is an important model parameter; reviewers disagreed about the most appropriate value. Differences in this parameter can result in appreciable variability in the modeled mercury concentrations for the human populations of interest.

Chapter 4 on toxic effects of various mercurials, particularly methylmercury, was the subject of several comments. The organization and presentation of toxic endpoints in the chapter could benefit by progressing from lethal through acute effects to subchronic and chronic effects. Distinct subsections organized along this framework would improve the presentation. The rationale for selecting the set of core studies of toxic responses should be clarified.

Not surprisingly, many comments involved the chapter on dose-response relationships. Several reviewers were concerned that the current RfD for methylmercury might not be protective, particularly for more subtle neurotoxic endpoints such as neurobehavioral and neurodevelopmental endpoints. One reviewer pointed out some confusion regarding the interpretation and presentation of the apparent association between maternal methylmercury exposure and abnormalities in deep tendon reflexes in their male children. Two reviewers recorded their disagreement regarding the adjustment of No Observed Adverse Effect Levels (NOAELs) and Lowest Observed Adverse Effect Levels (LOAELs) to lifetime exposures for different exposure pathways (e.g., inhalation, ingestion) in the derivation of RfDs and reference concentrations (RfCs). Exposure resulting from these pathways would be more realistically described as intermittent, shorter-term events. There was apparent confusion regarding the derivation and use of uncertainty factors (UFs) and modification factors (MFs). The values were not carried through the analysis according to the usual protocols. Reviewers pointed out some confusion and inconsistency regarding the relative sensitivity of adult and fetal developmental toxicity used to derive overall human health assessment endpoints.

Reviewers disagreed with the presentation regarding the possible interactions between mercury and selenium, particularly the implication that interaction with selenium may mitigate the human toxic effects of mercury.

3.2.3 Volume V (Ecological Effects) — Steven Bartell, Ph.D., and Paul Mushak, Ph.D.

Reviewers were concerned with the efficacy of the overall approach to the report's ecological assessment, which involves defining overlapping areas of potentially high mercury exposures with the distribution of sensitive piscivorous birds and mammals. For example, the life history and distribution of the Florida panther differ considerably from those of the mink or kingfisher. Failure to address life history and migration patterns in developing this overall approach might lead to inaccurate assessments of risk.

Reviewers also pointed out the report lacked a consideration of mercury effects on organisms at lower trophic levels (e.g., plankton, invertebrates). Additional reservations were expressed over the absence of wading birds, particularly species of declining abundance that are known piscivores. Effects of mercury on fish and reptiles should also be explored, or their omission should be further justified.

Reviewers were concerned about the report's dependence on assessment approaches and data that emphasized the Great Lakes and upper midwestern lakes, for example, in developing the bioaccumulation factors (BAFs). Concern also was expressed regarding the removal of surface waters with pH > 5.5 from regions of concern. This approach would exclude the circumneutral waters of the Florida Everglades that are suspected of posing mercury-related risks to resident populations of birds and mammals.

A major review issue focused on the use of NOAELs as endpoints for developing the wildlife criteria for the ecological assessment. This approach removes any consideration of a dose-response relationship from the assessment. If measured or modeled mercury exposures exceed the wildlife criteria values, we would not know the nature or magnitude of the expected response. Also, this approach implies different time scales between the shorter-term toxicity data used to develop the wildlife criteria and the longer-term exposure values. The fact that limited data were used to develop NOAELs for the selected wildlife species also calls into question the efficacy of the report's overall approach for estimating ecological risks.

In developing the BAF values, the report essentially ignored the complex chemistry of mercury in surface waters. Instead, these factors were developed using constant ratios of methylmercury to total water column mercury. Reviewers expressed serious concerns with this assumption, which ignores the complex environmental chemistry of mercury. Also, in developing the BAF values, the assumption was made that the selected piscivores restrain their feeding to specific "trophic level" fish. This assumption is certainly open to question; it remains unclear what the impacts of this assumption are on the resulting estimates of BAFs and wildlife criteria used as endpoints for the assessment. The assumption of a simple linear food chain implied by this approach was similarly of concern; the draft does not address spatial and temporal variations in diet and feeding behavior that might increase or decrease exposures for the selected piscivores.

It was not clear what the exposure models (RELMAP, COMPMERC) really provide to the assessment. The different spatial scales of these exposure models were not related to the spatial scale of the distributions of the selected species.

Finally, the reviewers noted that the sensitivity/uncertainty analyses did not comprehensively address all the components of the equations used to develop the BAFs or the final wildlife criteria values. The reported analyses addressed some of the models' structural uncertainties (e.g., correlations), but did not adequately address parameter uncertainty. The results of the sensitivity analyses do not lend themselves to defining future research needs in relation to reducing uncertainty on the endpoints of the assessment.

3.2.4 Volume VI (Risk Characterization) — Pamela Shubat, Ph.D., and Paul Mushak, Ph.D.

Reviewers agreed that Volume VI fell short of expectations for a risk characterization of health and ecological effects from mercury emissions. One reviewer felt that the necessary data to conduct a risk assessment are lacking, considering that a risk characterization should estimate the probability of health effects.

Reviewers noted that the volume should have compared the measurements of fish mercury levels and the incidence of health effects in populations to the volume's assumptions and results. The volume assumed a body weight and a fish consumption rate for each species; it also assumed a NOAEL and LOAEL for the selected species and derived a fish concentration that would permit consumption without exceeding the NOAEL or LOAEL. Reviewers felt that more data were needed to support this approach, and they expressed particular concern about the NOAEL and LOAEL selected for each species.

Reviewers felt that the assumptions, in the relative exposure ranking, that a given lake has only a single mercury concentration and a single trophic level were not accurate. The exposure rankings for the eagle, kingfisher, otter, and other species should be compared to measured values in tissue samples from these species.

3.3 Summary of Breakout Group Discussions

This section presents the summary of breakout group discussions on the following volumes of the Mercury Study Report to Congress:

- Exposure Breakout Group (Volumes II and III)—Gerald Keeler, Ph.D., and Paul Mushak, Ph.D; and
- Effects Breakout Group (Volumes IV, V, and VI)—Steven Bartell, Ph.D., and Paul Mushak, Ph.D.

3.3.1 Exposure Breakout Group (Volumes II and III)—Gerald Keeler, Ph.D., and Paul Mushak, Ph.D.

Volume II (Emissions)

Panelists suggested that the "minor sources"—i.e., those not included in the quantitative assessment—may contribute as much as an additional 20 percent to the total amount of mercury emitted annually. European emission factors should be used to improve the accuracy of this assessment of the minor sources.

Reviewers stressed that, to provide a complete picture of the atmospheric flux of mercury and to properly assess anthropogenic contributions to environmental mercury, the report should assess natural sources of atmospheric mercury as well as the reemission of mercury previously deposited on both aquatic and terrestrial environments by anthropogenic emissions.

Reviewers suggested that a national network of atmospheric mercury monitoring be established to validate emission data and to provide necessary information on trends in mercury deposition.

The panel felt that the division of sources into point and area source categories should be improved. For example, mercury emissions from residential heating furnaces should be defined as area sources, while crematories and medical waste incinerators should be categorized as point sources.

The panel agreed with the appropriateness of the emission factor approach. Many of the emission factors are based on actual test data and measurements, which contributes to the accuracy of the inventory. The emission estimates, when compared on a per capita basis, are quite similar to those in selected industrialized countries in Europe. In addition, the total U.S. anthropogenic mercury emissions are similar in magnitude to those of other industrialized nations in the world.

Volume III (Exposure Assessment)

The exposure volume utilized state-of-the-art methods in investigating the relationships between mercury emissions and exposures. Nevertheless, only plausible relationships between anthropogenic emissions and exposure could be defined.

The draft report does not assess the impact of anthropogenic mercury emissions in coastal environments. However, since fish consumption is the dominant exposure pathway, seafood or saltwater fish should be included in the total exposure estimates.

The analysis presented in the report supports the conclusion that current levels of emissions from major combustion/industrial sources result in incremental exposures above background to both humans and wildlife through the consumption of contaminated freshwater fish.

The group discussed the use of exposure estimates derived from the RELMAP and COMPMERC models. The discussants felt that the report should better describe how the model estimates were added. After questioning the modelers directly during the breakout group, the reviewers suggested that the authors consider alternative strategies for the risk assessment. For example, decoupling the regional impact provided by RELMAP from the local-scale exposure scenarios may improve the site-specific risk analysis and provide a clearer definition of the uncertainties in the exposure estimates utilized in the risk assessment.

Reviewers recommended that actual observations (i.e., measured mercury concentrations) could be used to "ground-truth" the model estimates or could themselves be used in the local-scale risk

assessments. Although a wealth of high-quality atmospheric mercury data or mercury deposition data is not available, enough data are available from the Great Lakes programs to perform a risk assessment at a similar or better level of accuracy than the models provided. The only drawback to this approach would be the lack of assignment of risk to specific source categories.

Additional suggestions for improving the assessments include:

- Evaluate the existing exposure to methylmercury via seafood consumption. Base this evaluation on existing data and not the model results.
- Perform the risk assessment and exposure to methylmercury from existing freshwater fish data. (This could be time-consuming because so many data are available.)
- Utilize existing wet and dry deposition data as input to the Indirect Exposure Model (IEM) to see what is predicted. This approach would remove two of the greatest uncertainties from the modeling and could be used to estimate the risk in the risk characterization.
- Attempt to identify a better indicator of the central tendency (perhaps the median) from the exposure assessment uncertainty analysis, which used the distributions rather than the high-end (maximum) estimates.

In conclusion, the panel members felt that the accuracy of the estimates decreases as the report moves from the initial emissions inventory through the exposure modeling using RELMAP and COMPMERC to the risk assessment phases. This results in a risk assessment that may have relatively large uncertainties and, therefore, may not provide a sound basis for decision- or policy-making.

The report would be improved by providing linkage between the risk management and the emissions inventory. The type and cost of mercury control technologies depend largely on the form of mercury in an emission and, thus, on the source category being considered for emission reduction.

3.3.2 Effects Breakout Group (Volumes IV, V, and VI) — Steven Bartell, Ph.D., and Paul Mushak, Ph.D.

Volume IV (Health Effects)

After some discussion, all or most group members generally agreed with the views and recommendations reported below. Dissenting views on key issues, where they occurred, are noted.

The group expressed several concerns about the organization and accuracy of Volume IV. Chapter 4 is difficult to follow, but group members generally agreed that its goal was to provide toxicity data for a human health risk assessment.

The description and discussion of lipophilicity of mercury compounds was not entirely accurate. The term is simplistic and does not account for current knowledge of binding and ligand-transfer interactions of methylmercury and other mercurials.

With respect to toxicity endpoints, the group noted that developmental impacts in the neonatal period should not be dismissed, since neonatal effects of elemental mercury have been reported in mice.

Differential sensitivity to mercurials among human populations is well established, and the fetus is now assumed to be the most sensitive to effects of methylmercury. The basis of such sensitivity includes physiological vulnerability, population variability concerning biotransformations (e.g., demethylation of methylmercury by gut flora), and variable patterns of exposure. Overall, sufficient data are not available to generate a highly resolved summary of differential sensitivity.

Of concern to the reviewers was treatment of the time course of exposure-effect relationships—i.e., are we dealing with latency or a masking phenomenon with long-term exposures?

Some reviewers were critical of the RfD calculation for inorganic ionic mercury (i.e., back-calculating from the drinking water equivalent level [DWEL]). Some also questioned how good a surrogate the Brown Norway rat is for humans sensitive for renal effects in the form of an autoimmune glomerulonephritis. One reviewer thought that the Integrated Risk Information System (IRIS) document is not convincing in this regard, and recommended that the mercury report at least reproduce the DWEL.

How UF factors were used in the analysis was not clear; the RfDs and RfCs need a closer look. Authors should reexamine the original data to see if they can justify how they used the numbers, and they should better explain their rationales.

The report should indicate that additional studies are under way (other than the Iraqi data set), although it is not known when the data will be available. Basically, the message here was to proceed with caution, but proceed.

Either Chapter 2 of Volume IV should be expanded to provide a concise summary of the integrated exposures to mercury, or an integrating final section should be added in Volume III. The authors should include more information on mercury exposure from dental amalgams and from ground waters that are or will be drinking water sources—particularly when mercury concentrations in these waters approach or exceed the RfC or RfD. Information should be added on how dietary components (other than methylmercury in fish) contribute to human exposure. This should include information, however qualitative, on any linkages of nonfish dietary mercury to atmospheric emissions.

Several comments concerned the mechanisms of mercury toxicology in humans and test animals. Although mechanisms of toxicity are critical to understanding the plausibility of epidemiological relationships reported for different populations and to understanding where thresholds for toxic effects may lie, the report gave them short shrift. The report should expand the discussion of this topic and should address how mercury forms move in and out of cells. However, reviewers recognized that a complete mechanisms sections might require an effort beyond the scope of the report.

Reviewers generally agreed that the health endpoints selected for the assessment and the dose-response relationship for each of the three forms of mercury were appropriate for the risk assessment. However, they thought the authors should strengthen the discussion of the validity of the endpoints and epidemiological data selected. Also, the group recommended that authors scrutinize the numbers employed from modeling, such as the fraction that goes into blood, the half-life, and the elimination parameter. The hair: blood ratio of 250 seems to be a middle-of-the road number and is probably acceptable. Reviewers questioned why the report did not use distributional analysis rather than selecting point values that might result in an unknown bias.

The group's comments on Appendix C of Volume IV mainly concerned model uncertainty and not variability in data-based parameters.

Reviewers considered the issue of selenium-mercury interactions. They felt this issue was complicated because the data sets are isolated and have no mechanistic underpinning. The critical question is how selenium in diet affects long-term exposures and associated chronic toxic endpoints. Was the Iraqi population at risk because of dietary habits (i.e., because they were grain eaters)? On the other hand, the reported selenium content of cereal grains is not vastly different than the selenium content measured in certain fishes. Although the selenium issue may have a bearing on which population exposed to methylmercury is valid for risk characterization, reviewers felt it premature to use selenium intake as a criterion for selection. One problem concerning the selenium-mercury connection is that the clearest associations are seen in gross endpoints, such as high-dose teratogenesis.

Regarding which dose-response data to use in risk characterization, reviewers expressed some sentiment for using at least two RfDs: one for the general adult population and one for pregnant women. Reviewers emphasized that the methylmercury RfD used in the assessment should be reported as an interim value, and that the assessment should be formulated to facilitate near-term (i.e., within the next several months) modifications to the RfD.

Some comments expressed in the effects breakout group also concerned the risk characterization volume. For example, the values of the NOAELs or LOAELs should be carried forth into the risk assessment instead of transforming them into permissible fish tissue concentrations.

Volume V (Ecological Assessment)

The group generally agreed that the goal was to provide data for a risk assessment and that the appropriate species were identified except for lower trophic levels and wading birds.

There was consensus that methylmercury was the compound of interest in addressing the toxic effects of mercury on piscivores. The consensus was further evidenced by the reported mortality of panthers, which was diagnosed as mercury toxicosis. The group also discussed the fact that the population of wading birds in the Everglades has significantly decreased in the last 5 years. Loss of habitat and exposure to mercury were listed as the suspected causes of these declines. One reviewer reported that loons in Minnesota also were suffering increased mortality from mercury exposure. Analyses showed elevated mercury concentrations in the feathers of juvenile loons. Approximately 2,500 loons died in coastal waters off Florida, in part from mercury exposure.

One reviewer pointed out that ethylmercury was measured in the Everglades, but this compound was not expected to be environmentally or toxicologically important in the overall assessment. Ethylmercury has not been identified in fish, for example. Dimethylmercury also exists in nature, but is quite volatile and, based on known information and the compound's fundamental chemistry, is not expected to represent any significant ecological threat.

Reviewers generally agreed that the report's treatment of methylmercury as a constant fraction of total mercury in the water column was an oversimplification. Additional work might be undertaken to determine the impacts of this assumption on the final estimates of the BAF and wildlife criteria values developed as assessment endpoints.

The group discussed the fact that chronic toxicity tests for methylmercury are extremely limited and that such effects are difficult to demonstrate under field conditions. For example, eggs can be collected from the nests of mercury-contaminated birds; however, it is not easy to detect toxic effects of mercury (e.g., hatching success, survivorship, growth). Different histories of exposure for adult birds may also make it difficult to establish effects in the field. As a result the reviewers suggested that the use of toxic effects measured in the laboratory is justified, particularly developmental effects. In other words,

laboratory-to-field extrapolations should be conserved. The group expressed concern about whether frank toxicity is the most appropriate endpoint, but acknowledged that frank effects are the best known.

A couple of reviewers thought that the dose-response relationships were adequately treated, the choice of a NOAEL and LOAEL was acceptable, and the limited toxicity data were used in an appropriate manner to develop the NOAELs and LOAELs used in the assessment. Some discussion ensued concerning the utility of toxicity data from laboratory studies on other animals (e.g., domestic animals and birds); these data might be used to at least help define the range of toxic exposure concentrations. The assessment needs to clarify the use of the wildlife criteria values developed in an approach paralleling human health risks (i.e., protection of individuals) for protecting populations of the selected wildlife species.

There was considerable discussion and concern regarding the validity of the overall conceptual model for the ecological assessment. This relates in part to the consideration of the complex chemistry of mercury in surface waters, where different physicochemical factors might determine exposure. Reviewers noted that lakes located side by side might show very different concentrations of mercury in fish. This multifactor complexity calls into question the linearity implied in the current approach for developing the BAF and wildlife criteria values. The concern is particularly important given the national scope of the intended assessment.

The reviewers noted the need to better articulate the uncertainty regarding the BAFs and the selection of the mean value. They also felt the report needed better discussions of distributions and of the nature of the uncertainty analysis.

Volume VI (Risk Characterization)

The effects breakout group's primary concern regarding Volume VI was its lack of emphasis on risk integration. Volume VI mainly reiterates and summarizes the material presented in the first five volumes. The reviewers were disappointed to find that the wildlife criteria values developed in Volume V were not carried directly through to the risk characterization. Substituting fish tissue mercury concentrations that are consistent with the wildlife criteria values is acceptable as long as the authors can clearly explain in the report why this was done. Nevertheless, the tissue concentrations (or, preferably, the wildlife criteria), should be developed as distributions, not single values. These distributions should be compared with distributions of expected mercury exposures on a regional basis for each of the selected piscivores. Such comparisons, which are more consistent with a probabilistic framework for ecological risk, will quickly identify species and regions of concern. They also will highlight where current information on exposure or toxic endpoints is insufficient to develop distributions that are precise enough for an assessment. Methods such as sensitivity and uncertainty analysis can then be used to examine the variance underlying such imprecision to pinpoint the major factors (e.g., model structure, model parameters) contributing to the overall uncertainty. Identifying the sources of uncertainty is important to promote efficient and effective allocation of limited resources and to improve precision, reduce bias, and refine the overall ecological assessment.

Reviewers felt the risk characterization might also address the risks posed by mercury to production dynamics at lower trophic levels. Clearly, such impacts have a profound effect on fish production that is independent of the direct accumulation and toxic effects on fish. These indirect effects are also relevant for assessing human and piscivore exposure to contaminated fish—fewer, smaller fish translates into reduced exposure, or at least a greater effort to obtain fish and, thus, significant mercury exposure if a larger number of smaller fish are consumed.

The group also expressed concern regarding the report's nearly total reliance on unverified models to produce the risk assessment. Where possible, the models that provided estimates of regional deposition and exposure should be evaluated in relation to known mercury concentrations. Any efforts at "ground-truthing" either the exposure or the toxicity models should be pursued within the resource and time constraints imposed by the overall schedule for delivering the report.

3.4 Overview of Reviewer Discussion in the Plenary Session — Paul Mushak, Ph.D.

Volume VI (Risk Characterization)

Panelists noted that a considerable portion of Volume VI consisted of summaries of Volumes II, III, IV, and V. These summaries covered human and wildlife health effects, overlay maps of sensitive wildlife populations with predicted high mercury depositions, and the uncertainties and assumptions in modeling emissions. Volume VI then provided a relative exposure ranking, a relative dose-response ranking, and levels of methylmercury in fish tissue that would be of concern for fish eaters.

The panel found the summaries to be confused and lacking; they failed to provide a comprehensive or quantitative discussion of the uncertainties and assumptions, and they did not discuss the extent and magnitude of the harmful exposures. Insufficient attention was given to linkages between anthropogenic emissions and background mercury data with the risk characterization.

One reviewer suggested that an ecological risk assessment be performed by using distributions of the parameters used to develop Tables 4-3 and 4-4 of Volume VI. Reviewers were impressed with the uncertainty analysis for the human RfD value found in Volume IV, Appendix C, and were interested in a discussion of propagated uncertainties.

The methodology and results in the comparative risk chapter of Volume VI were major areas of concern. Reviewers pointed out that the NOAELs and LOAELs are not based on the same set of endpoints and, therefore, are not directly comparable; in fact, the NOAELs and LOAELs may reflect a wide range of adverse responses. Another important concern was that the human NOAEL did not account for uncertainty areas such as different sensitivities. This indicates that use of the RfD would be more appropriate.

Regarding the wildlife criteria, reviewers felt that use of the published rat and monkey dose-response data would potentially capture more subtle effects in the rat. Notwithstanding the problems, information is available to enhance the accuracy of the criteria.

Reviewers offered several caveats regarding the strength of the linkages between point source emissions of mercury and increased levels of methylmercury in fish. Reviewers agreed there is no doubt that fish in certain areas exceed advisory limits. One reviewer claimed that all the conclusions in Volume VI are based on models rather than actual data. The volume would benefit from a discussion of the pathways for which there are claimed to be no data. Reviewers discussed the extent to which the report went beyond actual data, but did not come to a clear consensus.

In terms of fish consumption rates, reviewers felt the estimates of the distribution of such intakes should be improved.

Reviewers agreed that there is a significant need for systematic collection of data on increased levels of methylmercury in exposed wildlife populations.

In the aggregate, the discussion clearly indicated a need to better integrate the exposure and health effects data—for example, by comparing distributions of fish mercury levels with distributions of wildlife criteria. Some reviewers argued that background (baseline) determinations were needed to better determine increases over time. The panel also suggested that the RfD be clearly defined as "interim" and that it be revisited periodically as new data become available. Panelists also questioned the validity of comparing a human NOAEL to overt toxicity-based guidelines in wildlife, and why an RfD was not used.

Several comments concerned specific chapters in Volume VI. Deposition rates drive the overall analysis, and field verification is desirable. With reference to this, the exposure breakout group chair reemphasized that very recent data document the linkage between anthropogenic mercury emissions and deposition (e.g., the existence of a gradient with distance). Also, reviewers agreed that the report should better characterize seafood consumption, since it elevates the baseline for mercury exposure to which freshwater mercury intakes are added for the overall risk characterization. In addition, the panel recommended that seafood levels not be called "background" because some fraction of mercury in seafood is likely to come from anthropogenic sources.

Volume VII (Risk Management)

Reviewers agreed that Volume VII was generally good, but felt that it emphasized controls and did not adequately examine pollution prevention options. Pollution prevention could include banning products containing mercury (e.g., Minnesota's ban on mercury batteries). Reviewers also expressed concern about the volume's cost estimates for mercury control. For example, could the aggregate cost of reducing mercury emissions by half be calculated?

Reviewers thought it economically inaccurate to allocate all the costs of mercury reduction strictly to mercury, since typical reduction technologies also remove other contaminants. They suggested that the authors lower the cost estimate for mercury reduction by distributing reduction costs over all contaminants controlled by the technologies.

The panel felt that the absence in Volume VII of recommended actions and research needs is a major gap that should be filled. Recommendations could include, for example, market-based approaches, product reformulations, product bans, and recycling. The European experience was suggested as a valuable source for information on market-based approaches.

3.5 Summary of Major Revisions Made in Response to Reviewers Comments

All volumes:

- Executive summaries re-written to be more informative
- Executive summaries written to include conclusions categorized by degree of confidence in the findings, summaries of uncertainties and research to improve the assessment.

Volume II: An Inventory of Anthropogenic Mercury Emissions in the United States

- Revised natural emission inventory information to be consistent with Expert Panel Report.
- Added industrial use trends and historical trends
- Updated municipal waste combustor (MWC) inventory to include 50 closures; this resulted in a decrease in the emissions estimate of 10 metric tons/yr to 55 metric tons.
- Added impacts of proposed medial waste incinerator (MWI) and MWC rules.
- Revised inventory to use 1993 instead of 1992 Bureau of Mines data.
- Incorporated maps showing locations of sources.
- Incorporated industry-specific comments.

Volume III: An Assessment of Exposure from Anthropogenic Mercury Emissions in the United States

- Numerous recent peer-reviewed studies were incorporated.
- Sections added on exposure from anthropogenic , non-ambient sources including dental amalgam, occupational exposure and consumption of marine fish.
- Section added on measured mercury concentrations near multiple local sources
- Additional mercury measurement data from various media added and compared to modeled estimates. These measurements included air concentrations, deposition rates and soil concentrations.
- An assessment of the mercury exposures that result from the input of measured mercury air concentration, deposition rate and soil concentration data to the indirect exposure models was added.
- Modeling assumptions were modified to accommodate new data.
 - Increased percentage of divalent mercury assumed to be particulate bound.
 - Flat terrain only was modeled and effects of complex terrain addressed separately in an uncertainty analysis.

- The configuration of the watershed was changed and area-averaged deposition rates were utilized.
 - The aquatic trophic levels, which wildlife were assumed to consume, were modified.
 - The assumed quantity of background atmospheric mercury was modified.
 - Deposition velocities for vapor-phase divalent mercury were modified to account for lower dry deposition rates at night.
 - The assumption related to the bioconcentration of atmospheric mercury into green plants was modified to account for lower measured concentrations in edible portions of grains and legumes.
- Exposure models were re-run to accommodate the above assumptions and the revised emissions inventory.
 - A section (Appendix H) was added to estimate the size of the fish consuming human population in the U.S., the amounts of fish consumed by the general U.S. population and several high-end-fish-consuming populations, and the amount of mercury measured in surveys of marine and fresh-water fish. These data were used to generate estimates of mercury exposure from fish consumption. These mercury exposure estimates were not attributed to individual sources or source categories.

Volume IV: Health Effects of Mercury and Mercury Compounds

- Section added on pharmacokinetic models. No pharmacokinetic model was chosen for use in the health assessment.
- Germ cell mutagenicity assessment was re-written to remove the numbered classification.
- Additional studies on developmental toxicity of elemental mercury were added raising the overall weight of evidence judgement to “Sufficient Animal Data” for developmental toxicity
- The newly verified RfD for methylmercury was described. A section on input data and derivation of the benchmark dose was added as was discussion of plausible alternatives to the U.S.EPA RfD.
- Section on interactions of other materials with mercury and section on selenium were re-written.
- A section on mechanism of action of mercury was eliminated.

Volume V: An Ecological Assessment for Anthropogenic Mercury Emissions in the United States

- Discussion of several new studies supplied by the reviewers was added.
- Enhanced discussion of non-mammalian, non-avian life forms

- Obtained original doctoral dissertation describing effects in mink and used as basis for reevaluation of mammalian wildlife criteria.
 - Used revised no observed adverse effect level from dissertation
 - Described uncertainty factor of 10 for subchronic to chronic extrapolation
- Re-evaluated criteria for avian species
 - described available data on loons, but did not calculate a wildlife criterion for this species
 - Described studies from the National Biological Service on levels of mercury in eagle feathers.
 - Describe uncertainty in LOAEL to NOAEL extrapolation and species extrapolation
- Analyzed data from laboratory animal studies to bound uncertainty on wildlife criteria
- Clarified assumptions, uncertainties and methods in development of wildlife criteria.
- Described variability and uncertainty in wildlife feeding habits.

Volume VI: Characterization of Human Health and Wildlife Risks from Anthropogenic Mercury Emissions in the United States

- Volume was completely re-organized to meet specifications of new U.S.EPA guidance on risk characterization
- Discussion of plausible alternatives to the U.S. EPA RfD on methylmercury included.
- Revised and expanded discussion of uncertainty and variability
- Included estimates of size of “at risk “ human and wildlife populations
 - Human estimate based on data from National Center for health Statistics (CDC), U.S. census data, and the Continuous Survey of Food Intake by Individuals. This was combined with measured levels of mercury in marine and fresh-water fish.
 - Wildlife estimates made from literature.
- Highlighted exposure as the major source of variability vs. Species -specific differences in susceptibility to toxic effects.
- Added comparison of mercury exposure estimates with methylmercury RfD or equivalents for humans and wildlife.

Volume VII: An Evaluation of Mercury Control Technologies and Costs

- Enhanced discussion of pollution prevention opportunities. These were discussed in qualitative terms and quantified when data were sufficient.
- Integration of control costs with benefits was done, as well as final section on management alternatives and statutory authorities.

4. SUMMARY OF INTERAGENCY REVIEWERS COMMENTS AND DISPOSITION

Reviews of the External Review Draft of the Mercury Study Report to Congress were obtained from the following U.S. government agencies:

- Agency for Toxic Substances and Disease Registry (ATSDR), Public Health Service, U.S. Department of Health and Human Services.
- National Institute of Environmental Health Sciences (NIEHS), National Institutes of Health
- National Oceanic and Atmospheric Administration (NOAA), Department of Commerce
- U.S. Department of Agriculture (USDA)
- U.S. Department of Energy (DOE)
- U.S. Food and Drug Administration (FDA)
- National Biological Service

A meeting of reviewers was held at the U.S. EPA, Washington DC on 1/9/95 to discuss scientific issues concerning the report. Representatives of the above Agencies attended the meeting with the exception of NIEHS; comments from NIEHS were submitted in writing after the meeting. Written comments were requested of all reviewers; responses were received from ATSDR, DOE, NIEHS, USDA and the National Biological Service. At the meeting and in written reviews point of congruency among Federal risk estimates and methodologies were identified; points of divergent opinion were also noted.

Major critiques are described below as well as U.S. EPA's response (in italics). It is the Agency's intent to describe in the final Report alternate points of view or risk estimates in those instances wherein U.S. EPA disagrees with another federal agency.

General Comments on the Report.

- Reviewers noted that some references were incomplete or missing.

These were completed. To the extent possible within deadlines, papers submitted by the reviewers were cited in the Report.

- Reviewers felt that the Report would be greatly enhanced in its usefulness if general conclusions on the extent of mercury contamination or degree of hazard could be articulated in plain language.

This was done for inclusion in Vol. I Executive Summary, which was prepared after the interagency review was completed. In addition, each volume was revised to include a general conclusions summary in its own executive summary.

- Reviewers discussed section 112(n)(1) of the Clean Air Act Amendments of 1990. This specifies the following.

The Administrator shall conduct, and transmit to the Congress not later than 4 years after the date of enactment of the Clean Air Act Amendments of 1990, a study of mercury emissions from electric utility steam generating units, municipal waste combustion units, and other sources, including area sources. Such study shall consider the rate and mass of such emissions, the health and environmental effects of such emissions, technologies which are available to control such emissions, and the cost of such technologies.

- FDA proposed that U.S. EPA was not required to determine or comment upon a threshold for adverse effects of mercury in humans and that it was inappropriate for U.S. EPA to make such a determination in this Report.

U.S. EPA is obliged to follow consistent methodologies and published Guidelines for Human Health Risk assessment in its evaluation of potential human health impacts of environmental agents. For general systemic non-cancer health endpoints this includes consideration/calculation of reference doses (RfD) or reference concentrations (RfC). The methods used for derivation of RfDs and RfCs are based on the assumption of a population threshold for response in the absence of data which indicates that no threshold exists. It was agreed by both scientific staff and U.S. EPA management that application of state-of-the art methodologies for calculation of RfDs and RfCs was an appropriate part of the Mercury Study. A statement of the FDA critique is included in the section of the Report summarizing reviewer comments.

- Reviewers noted that the Report did not deal with the impacts of global mercury use or emissions or of "natural" mercury.

U.S. EPA was directed in the CAAA to deal with emissions from various specified sources and "other sources, including area sources". When data were sought and models constructed, it became obvious that contemporary, reliable emissions data on mercury were not sufficient to support a national survey. Neither the extant data nor modeling technology permitted accurate modeling of emissions from countries other than the U.S. The acknowledged global cycling of mercury was accounted for in the incorporation of a 1.6 ng/m³ "background level" into the long-range transport modeling (RELMAP). The Report describes the impossibility of determining whether mercury is of "natural" or anthropogenic origin; there is, for example a discussion of hypotheses that mercury soil levels in sites distant from emissions sources can be the consequence of deposition over time of mercury released as a result of human activities. The Executive Summary and Exposure volumes indicate that any local evaluation of mercury hazard must use local determinations of mercury in media.

Volume II: Inventory of Anthropogenic Mercury Emissions in the United States.

- The emissions inventory was thought generally to be comprehensive and well described. There was general agreement with the conclusions on relative source contributions.
- An explanation should be given in this volume of the use of emissions data in the exposure modeling.

This was done.

- A description of derivation of emission modifying factors (EMF) was requested; specifically, were these numbers means, representative values, etc.

This information was added.

- USDA proposed the lack of an estimate of mercury emissions from landfills was a serious deficit.

Data are not available which permit any sort of generalization about the magnitude of emissions from this source type. There is only one study of mercury emitted from a landfill area; this was done in Minnesota, and there is no indication that this site was representative of other waste sites. Studies on landfills as a potential mercury source have emphasized pathways leading to groundwater contamination rather than release to the air.

- USDA also remarked that mercury from application of sewage sludge to farm land was not considered as a source.

The Report does include some information on sewage sludge incineration and its potential for mercury release to the atmosphere. Data on consequences of land application of sludge, to the extent provided by the USDA, were included in Vol II or III as appropriate.

- The National Biological Service recommended adding more information on the re-emission of deposited mercury of anthropogenic origin.

This is discussed in the Report as a source for which data are not available and as a contributor to possible underestimation of over all emissions.

Volume III: An Assessment of Exposure from Anthropogenic Mercury Emissions in the United States.

- There was general agreement that mercury deposition model results were reasonable predictions given available data. There was discussion of impacts of using emission factors from washed coal and from seams most recently used by coal fired utility boilers on the relative ranking of source contributions. There was discussion of the likelihood that methylmercury is released from utility stacks; consensus opinions of U.S. EPA and reviewers were that there were no conclusive data on methylmercury release.

- USDA raised concern that parameters (e.g., amounts of foodstuffs consumed by human populations) used in Vol III modeling were inconsistent with those used in the sludge evaluations.

The parameters were compared, and any departures from the sludge methodologies are described; justifications for departure are provided.

- USDA identified consumption of wild mushrooms as a source of mercury.

Description of this source based on material if provided by USDA was included in Vol III.

- ATSDR noted that use of the term "subsistence fisher" in the assessment was inaccurate because the consumption rate used was not sufficient to constitute dietary subsistence.

Use of "high end fish consumer" or some other more descriptive term was substituted.

- DOE and others cautioned against using "significant" outside a statistical context.

Another term was used when statistical significance is not being described.

- There was discussion of the availability and usefulness of mercury total body burden data.

It was agreed to incorporate such data as were available from ATSDR and on recent reports from Sweden and on a group of Chippewa Native Americans. The Report describes the limitations on comparison of the modeled predictions with body burden data. Body burden data include cumulative exposure to non-anthropogenic mercury and mercury in marine fish.

- There was discussion on variability in estimates of percent of mercury in food sources as methylmercury.

The Report describes this variability. Sources of the estimates were checked to ensure that attribution is clear.

- There was a brief discussion of the impact of dental amalgams on total mercury body burden.

Discussions in the report on amalgam mercury release were reiterated in the beginning of Vol III in the section outlining those sources which were modeled, how background is considered, etc.

- Several reviewers pointed to the lack of information on marine fish. It was noted by FDA that one cannot generalize as to whether marine fish or freshwater fish are likely to have higher concentrations of mercury.

The modeling of mercury deposition employed by U.S. EPA of necessity dealt only with mercury in U.S. continental, fresh water lakes and streams. The Report contains one table on measured mercury levels in commercial marine fish. This was enhanced with data supplied by the reviewers, and the accompanying discussion was expanded and moved to the beginning of the report. Conclusion statements of the Report acknowledge that the majority of fish consumed in the U.S. is marine fish. Marine seafood consumption estimates are included in discussions in Volume VI.

- The NOAA and the National Biological Service reviewers said that the mercury species found in fish flesh varies with the type and trophic level of fish.

Variation reported in the literature is described in Volume III.

- The reviewer from the National Biological Service took issue with some assumptions used in the deposition modeling; specifically, he asserted that most precipitated mercury is particulate bound and that methylmercury can be introduced into systems by wet deposition.

The recommended papers (Benoit, Fitzgerald and Damman, 1994., Holtberg et al., Monterey Conference Proceedings) were evaluated as to inclusion in the Report. Note that Benoit et al. is in press and was not available for evaluation in the time frame of U.S. EPA's Report deadlines.

- The National Biological Service registered a strong objection to the use of bioaccumulation factors or other means to make generalized statements as to relationship between mercury in water and predicted concentrations of mercury in fish inhabiting the water. The reviewer stated that local biogeochemistry is highly variable with the result that fish taken from water bodies with the same mercury concentration can have very different mercury concentrations in the tissue. Discussion focussed on factors governing this variability; it was acknowledged that there are no data to allow modeling of any one factor or combination of factors. There was discussion of use of the EPRI Mercury Cycling Model (MCM). There was agreement that this is not appropriate as a basis for local or general conclusions as to relationship between water and fish mercury concentrations. The objection to the MCM stems in large part from its basis on data from a water body not considered to be representative of other U.S. freshwater lakes.

U.S. EPA maintains that some form of estimate of fish tissue mercury level is needed to evaluate the potential impact of anthropogenic mercury emissions on human and wildlife health. The Report was amended to include local biogeochemistry as a source of substantial variability in fish mercury predictions. Ranges of mercury fish levels provided by the National Biological Service were used to describe the extent of variability. This was added to the discussion of limitations of use of the modeled estimates for any site-specific evaluation.

- USDA described the potential for sheep to consume beet greens which may be a source of mercury.

Volume III was reviewed to ensure that sources of mercury contamination not modeled are mentioned.

Volume IV: Health Effects of Mercury and Mercury Compounds.

- There was agreement with the hazard identification categories for carcinogenicity, developmental toxicity and germ cell mutagenicity for elemental, inorganic and methylmercury with the exception noted below. It was agreed that no low dose extrapolation for potential carcinogenicity of inorganic mercury or methyl mercury is supported by existing data. It was agreed that immune-mediated glomerulonephritis is the critical effect for a reference dose for inorganic mercury. It was agreed that a reference concentration for elemental mercury of 3×10^{-4} mg/m³ is reasonable. After much discussion (excerpted below) it was agreed that a reference dose for methylmercury is within an order of magnitude of 10^{-4} mg/kg-day.
- It was recommended that "methylmercury" be substituted for "organic mercury".

This change has been made in the Report.

- U.S. EPA discussed the pending revisions to the Guidelines for Risk Assessment of Carcinogens.

While the alphanumeric classification was maintained in the text, discussion of these classifications was enhanced to conform to the narrative classifications which U.S. EPA will likely use in the near future. In addition the number classification in the discussion of germ cell mutagenicity was dropped.

- ATSDR indicated that there are new data on developmental effects resulting from inhalation exposure to elemental mercury.

These studies were evaluated and the classification of "insufficient evidence for developmental toxicity" re-examined.

- In its derivation of an intermediate MRL for inorganic mercury (2×10^{-3} mg/kg-day) ATSDR used a NOAEL 0.23 for F344 rats gavaged for six months as part of the subchronic range-finding component of a cancer bioassay (NTP, 1993).

This study was not available at the time that U.S. EPA convened an expert panel to derive its RfD for inorganic mercury. That panel recommended use of data from short term studies in Brown Norway rats as an animal model appropriate to estimation of potential toxicity in sensitive human subpopulations. The NOAEL and LOAEL from the NTP bioassay are within the range observed in three studies in the Brown Norway rat. U.S. EPA scientists have concluded that the existing RfD, described in the 1988 Drinking Water Criteria Document for Inorganic Mercury is not impacted by the more recent data from NTP. The ATSDR evaluation is described and compared to U.S. EPA's in the risk assessment chapter of Vol IV.

- The FDA reviewer stated that in deriving an RfD for methylmercury (and other agents) U.S. EPA does not estimate or predict the degree of risk but rather estimates a measure of a "safe" level of exposure. The reviewer felt that the "bright line" approach does not constitute a risk assessment.

There was some agreement with the reviewer's position, particularly in the utility of predicting risk above a hypothetical threshold. U.S. EPA, however, has not completed analyses which would support such an estimate of risk. The question of whether the data used (neurologic deficits in children of Iraqi mothers who ingested contaminated grain during gestation, Marsh et al 1987) are suitable to this type of analysis is an open one. At this time U.S. EPA does not include any estimate of risk above the RfD in the Report. Discussion will be continued by U.S. EPA and FDA scientists with the goal of deriving an estimate of methylmercury risk for ingestion levels.

- There was discussion of the impact of current studies of developmental effects in populations which consume high end levels of marine fish and/or mammals (the Faroe Islands and Seychelles Islands studies). Some results these epidemiologic investigations have been presented at recent meetings and have been published in abstract in proceedings. It was the opinion of the FDA reviewer that these studies show no (or little) neurologic impairment in children exposed *in utero* to mercury levels associated with observed effects in the Iraqi population on which U.S. EPA based its RfD. U.S. EPA was encouraged to use these data in their quantitative assessment of non-cancer effects.

U.S. EPA can only use data which are available to the scientific community and have undergone a process of peer review. The deadlines specified in the CAAA do not permit delay until the studies have been published in the peer reviewed press or the data submitted to U.S. EPA for a process of expert review. (It was noted that U.S. EPA has missed the submission date (11/94) specified in the CAAA.) The Faroe and Seychelles Islands studies as reported in abstract are described in Volume IV. In response to the critique that there has been no influence of these results in U.S. EPA's risk evaluation, the Report was amended in the following ways. In both Vol IV and Vol VI (Risk

Characterization), the potential for the Faroes and Seychelles results to decrease uncertainty in the RfD is described. Alternative approaches are described; specifically, decreasing the uncertainty factor or using the upper bound on the 10% risk level for the benchmark dose (vs the lower bound which U.S. EPA employed). These alternatives are used to describe the range around the U.S. EPA RfD of 1×10^{-4} mg/kg-day.

Volume V: An Ecological Assessment for Anthropogenic Mercury Emissions in the United States.

- There was agreement that data are insufficient for evaluation of mercury impacts on any ecosystem. There was agreement that data were insufficient to calculate a wildlife criterion for Florida panthers. There was no objection to development of wildlife criteria for methylmercury only. It was agreed that lack of data on sensitive indicators of toxic effect in wildlife species is a major contributor to uncertainty in the estimates.
- The National Biological Service reiterated its concern with use of any method (such as a BAF) which relates water concentrations of mercury to fish concentrations.

Volume V repeats discussions of variability in fish concentrations due to local biogeochemistry.

- USDA felt that in derivation of the trophic level 3 and 4 BAFs that a geometric mean was more appropriate than the simple mean used.

All calculations were performed on the logs of values; arithmetic values presented in tables were converted from logs after derivation of means and percentiles. Geometric means were, thus, used in derivation of the BAFs. Estimates are included in description of BAF derivation.

- The reviewer from the National Biological Service objected to the presentation of maps showing overlay of wildlife habitat with mercury deposition and low and normal pH water bodies. The reviewer felt that the maps (with the Florida panther as the example) were misleading and gave a false impression that no problem exists for some species.

The maps were designed to show only predicted high mercury deposition and do not rule out the likelihood of mercury contamination in areas (particularly wetlands) contiguous to high deposition areas. The purpose of the overlay procedure was to highlight areas and species of concern, not to eliminate areas as of no interest. The extent to which overlap can be quantified is being examined; results will be included in the Report as feasible. The purpose and limitations of the overlay maps has been explicated more completely in Vol V.

- ATSDR indicated in the derivation of wildlife RfDs and criteria that interspecies extrapolation not based on pharmacokinetic data will have an unacceptable degree of uncertainty.

Thus far, no useful data on pharmacokinetics in the species of interest have been available. Additional literature searches are being conducted in that area. U.S. EPA scientists feel that an adjustment of the NOAEL reported for mink is not need for application to otters. The adjustment of the NOAEL derived in mallards for application to three fish-eating birds will be re-evaluated if data permit.

- Several reviewers queried whether the wildlife criteria were conservative. Questions were raised about the likelihood that wildlife have evolved protective adaptations to mercury toxicity.

Data are insufficient to answer either question. The endpoints tested in the wildlife species are neither as sensitive nor as subtle as those detected in humans exposed to methylmercury. There is no indication whether individual species or ecosystems are being impacted by mercury such that viability or reproduction is reduced. Discussion of this uncertainty will be expanded and reiterated in Volumes V and VI. Information from the National Biological Service on correlation between eagle feather mercury levels and reproductive rates were included.

Volume VI: Characterization of Human Health and Wildlife Risks from Anthropogenic Mercury Emissions in the United States.

- After much discussion there was agreement that data (limited as they are) for wildlife and humans do not show special sensitivity of one species over the others. The range of (adjusted) NOAELs is in within an order of magnitude.
- There was much discussion on the comparisons made at the end of Volume VI: NOAELs and LOAELs for human and wildlife populations, levels of mercury in fish which would result in exposure to NOAELs or LOAELs given assumptions of fish consumption. The utility of this approach was questioned by some reviewers; the soundness of the data and extrapolations were questioned by others.

U.S. EPA is reconsidering the comparisons made. Our preference at this time is for some form of interspecies comparison; an holistic approach to assessment of risk for human and non-human species is the direction which ORD is taking, based on recent mandates and advice to U.S. EPA. The method of comparison used in the Report is untried. It may be advisable to limit the scientific uncertainty by backing up a few steps; that is to limit comparisons to LOAELs and NOAELs without the additional step of including exposure assumptions to calculate reference levels of mercury in fish. The advantage of the last step is that it makes clear the relationship between measured adverse endpoint in species of concern and guidance levels such as fish advisories.

- Several reviewers found they could not follow the process of wildlife NOAEL estimation from the text or tables in Volume VI.

The estimation of all NOAELs and LOAEL is explained more fully in Volume VI. The use of uncertainty adjustments as proposed in the Great Lakes Initiative is explained.

- Reality checks as to measured levels of mercury in wildlife were requested by reviewers.

Information from Vol III, Vol V and new information supplied by the National Biological Service (e.g., levels of mercury in feathers) is carried over to Volume VI.

- There was agreement among all parties that the Report and Volume VI in particular should present conclusions as emphatically and clearly as the science permits.

Conclusions for all volumes are articulated and presented in each Executive Summary chapter. These conclusions are re-stated in Volume VI (for risk assessment) and in Volume I (for all conclusions).

Volume VII: An evaluation of Mercury Control Technologies, Costs and Regulatory Issues.

- There was agreement that the description of control technologies and the costs of controls was comprehensive and as accurate as extant data permit.
- Reviewers discussed the "societal cost" chapter of Vol VII. DOE asked whether a cost/benefit analysis was done. Reviewers asked if impacts on international trade (eg GATT) were considered. FDA inquired specifically if benefits of fish consumption (health and societal) were weighed against costs.

The CAAA mandate did not specify a cost/benefit analysis for this report. The study included only material which could be used for cost/cost comparisons (e.g., cost of mercury control vs. loss of revenue from recreational fishing). It was agreed after discussion that (unlike the situation for lead exposure) there are insufficient population data or economic impact data for subtle health effects to permit a suitable cost/benefit analysis.